Vertical Particle Concentration Profiles Around Urban Office Buildings

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Abstract

Despite its role in determining both indoor and outdoor human exposure to anthropogenic particles, there is limited information describing vertical profiles of particle concentrations in urban environments, especially for ultrafine particles. Furthermore, the results of the few studies performed have been inconsistent. As such, this study aimed to assess the influence of vehicle emissions and nucleation formation on particle characteristics (particle number size distribution - PNSD and $PM_{2.5}$ concentration) at different heights around three urban office buildings located next to busy roads in Brisbane, Australia, and place these results in the broader context of the existing literature. Two sets of instruments were used to simultaneously measure PNSD, particle number (PN) and $PM_{2.5}$ concentrations, respectively, for up to three weeks at each building.

The results showed that both PNSD and $PM_{2.5}$ concentration around building envelopes were influenced by vehicle emissions and new particle formation, and that they exhibited variability across the three different office buildings. During nucleation events, PN concentration in size range of < 30 nm and total PN concentration increased (7 – 65% and 5 – 46%, respectively), while $PM_{2.5}$ concentration decreased (36 – 52%) with height.

This study has shown an under acknowledged role for nucleation in producing particles that can affect large numbers of people, due to the high density and occupancy of urban office buildings and the fact that the vast majority of people's time is spent indoors. These findings highlight important new information related to the previously overlooked role of particle

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formation in the urban atmosphere and its potential effects on selection of air intake locations and appropriate filter types when designing or upgrading mechanical ventilation systems in urban office buildings. The results also serve to better define particle behaviour and variability around building envelopes, which has implications for studies of both human exposure and particle dynamics.

Keywords: Ultrafine particle, particle number size distribution, particle number, $PM_{2.5}$, building envelope.

1 Introduction

Epidemiological research has consistently shown an association between fine (< 2.5 μm; $PM_{2.5}$) particle concentrations and increases in both respiratory and cardiovascular morbidity and mortality (Pope, 2000; Davidson et al., 2005; Schwartz and Neas, 2000). The health effects of ultrafine (< 0.1 μm) particles are less well known, however research to date indicates that they may be equally or more detrimental than those of $PM_{2.5}$ and PM_{10} (Oberdorster, 2000; Franck et al., 2011).

Ultrafine particles make only a minor contribution to particle mass (~10%), but often constitute up to ~90% of particle number (PN), with these figures being reversed for fine particles (Morawska et al., 2008). The amount of fine and ultrafine particles in the urban atmosphere is mainly influenced by vehicle exhaust emissions during the traffic peak hours (Pey et al., 2008; Perez et al., 2010) and new particle formation by photochemical reactions (Pey et al., 2009).

Outdoor particles can penetrate the building envelope via doors, windows, building structure leakages, and especially via mechanical ventilation systems. It is therefore important to understand the vertical profiles, concentrations and dynamics of particles around the envelope in order to locate the optimal position for outdoor air intakes, and best mitigate the penetration of particles indoors. Moreover, such information is relevant to developing a better

1 understanding of the complex nature of particles in urban street canyons and their relationship
2 to pedestrian exposure at ground level.

3 To-date, studies investigating vertical profiles of particle mass concentrations around building
4 envelopes has yielded inconsistent findings. Some research concluded that concentrations
5 decreased with increasing height, including Horvath et al. (1988) who showed that diesel
6 particle mass concentration decreased by 17% at 27 m compared to street level. Micallef and
7 Colls (1998) found that PM_{10} and total suspended particle (TSP) concentrations at a height of
8 0.8 m above the ground floor were about 35% higher than those at a height of 2.9 m, while
9 Rubino et al. (1998) reported a decrease in the concentrations of PM_{10} with increasing height,
10 and the concentration on the leeward side of the building was consistently lower than on the
11 windward side. Chan and Kwok (2000) also found that the relationship between decreases in
12 particle mass concentrations and height was exponential in a street canyon and linear for open
13 sites. However, other studies have shown a decrease in particle mass concentrations to certain
14 heights, with concentrations remaining somewhat constant beyond that. In particular, Chen
15 and Mao (1998) reported that the PM_{10} concentrations on the seventh and fourteenth floors
16 were comparable, after sharply decreasing from the second floor to the seventh floor.
17 Additionally, Kalaiarasan et al. (2009) found that $PM_{2.5}$ concentrations were highest around
18 the mid-floors when compared to those measured at the upper and lower floor of high-rise
19 buildings. Bullin et al. (1985) reported a vertical TSP profile was nearly flat.

20 In contrast to particle mass, only a handful studies have measured PN concentrations around
21 the building envelope. Vakeva et al. (1999) monitored PN concentrations at street and rooftop
22 levels, and showed that the concentrations at 1.5 m were significantly higher than those at 25
23 m. Hitchins et al. (2002) also observed a decrease in PN concentrations with height when
24 measured at the front of a high rise building 80 m from road, but this was the opposite when
25 measured at the rear of this building. Longley et al. (2004) noted that total number
26 concentrations at 17 m were generally half of those at 4 m during the day and the gradient was

reduced significantly at night when measurements were conducted in an asymmetric street canyon. Similarly, Kumar et al. (2009) found that PN concentrations at street level (0.2-2.6 m high) were about 6.5 times higher than those at rooftop height (20 m). Other research conducted by Li et al. (2007) showed that PN concentrations decreased by 72 % and 85 % at a height of 38 m compared to that at 1.5 m when the wind blew parallel and perpendicularly the street canyon. Vakeva et al. (1999), Li et al. (2007) and Kumar et al. (2009) also discussed the influence of the photochemical aerosol particle formation relative to local vehicle emissions on vertical profile of PN concentrations. However, not only the local emissions but also other air mass from different regions, travelling with the wind direction can influence new particle formation in urban areas (Stanier et al., 2004; Qian et al., 2007; Hussein et al., 2008; Salma et al., 2011; Cheung et al., 2011).

In addition to research surrounding building envelopes, some studies have quantified the vertical profiles of particle concentrations in urban areas. Imhof et al. (2005) has shown that PN concentrations 60 m downwind of a highway decreased when measured at heights of 5 – 30 m. Zhu and Hinds (2005) quantified the vertical particle concentrations measured 50 m downwind of an elevated highway and reported that the PN concentrations increased within the first 5m from the ground, then decreased at higher levels. He and Dhaniyala (2012) measured vertical profiles of PN concentrations at heights between 0.55 to 10 m at distances 15, 50, and 100 m from a highway. Their results have shown that vertical profiles of particle concentrations vary with wind speed, direction and distance from the highway.

A relationship between PN and particle mass concentrations has also been reported for urban background sites, as well as in street canyons. For example, Harrison et al. (1999) found a significant linear correlation between PN and PM_{10} concentrations at an urban background location (R^2 = 0.44). Similarly, Longley et al. (2003) determined that the linear correlation (R^2) between ultrafine PN and $PM_{2.5}$ concentrations in a street canyon was 0.51. However, there may be a difference in correlations between particle number size distribution (PNSD)

and particle mass concentration around a building envelope due to the influence of different factors, such as emission sources, building height, and especially, the difference in particle size ranges.

Due to the inconsistent findings of previous studies, there is a lack of clear knowledge regarding PNSD, the factors affecting it, and its relationship with particle mass. The characteristics, variability and role of particle vertical profiles in both indoor and outdoor human exposure in and around urban buildings remains poorly understood. To contribute towards addressing these knowledge gaps and inform the limited experimental evidence base currently underlying numerous modelling studies, we aimed to: (1) assess the variation of PNSD, PN and $PM_{2.5}$ concentrations by simultaneous measurements at the rooftop and street levels of three urban office buildings; (2) quantify vertical profiles of PNSD and $PM_{2.5}$ concentration and analyse the influence of vehicle emissions and nucleation events on these vertical profiles; (3) quantify and interpret differences between PNSD and $PM_{2.5}$ concentration at different levels; and (4) place the results in the context of broader literature and seek to identify if location-independent trends exist for vertical profiles of PN and $PM_{2.5}$.

2 Experimental methods

2.1 Setting

Our research was conducted in the subtropical city of Brisbane, which is the capital city of Queensland, Australia. Detailed information on the topography and meteorology of this region is described in Cheung et al. (2011). The major air pollution sources found in the Central Business District (CBD) are inner-city traffic emissions, and aircraft, ship and industrial emissions transported from the lower reaches of the River, located approximately 15-18 km NE of the CBD.

We selected three urban office buildings, located close to busy roads with different terrains. Building A is ~17 m high, located on relatively flat ground with unrestricted access and ~7 m

from a busway, which is a bus-only roadway with a daily traffic volume of about 900 buses. Building B is ~77 m high, located in the centre of the CBD and surrounded by other high rise buildings and busy city roads with a daily traffic volume of about 11,000 vehicles. Building C is ~25 m high, located ~7 m from a freeway with a daily traffic volume of about 110,000 vehicles. There are some high rise buildings to the rear of this building.

2.2 Instrumentation

Two TSI 3934 Scanning Mobility Particle Sizers (SMPSs) were used for measuring PNSD in the range 8.5 – 400 nm. Each SMPS is comprised of a TSI 3071 Electrostatic Classifier (EC) that classifies particles according to their electrical mobility, and a TSI 3010 Condensation Particle Counter (CPC). The duration of each scan was 180 seconds. The PN concentrations in the range 6 – 3000 nm were measured using two TSI 3781 CPCs at an averaging interval of 10 seconds.

Two TSI 8520 DustTrak aerosol monitors, each with a 2.5 μm inlet were used to measure $PM_{2.5}$ concentrations at an averaging interval of 30 seconds. It should be noted that the DustTrak operates based on a light scattering technique where the amount of scattered light is proportional to the volume concentration of the aerosol. The DustTraks used to measure $PM_{2.5}$ concentrations in this study were not calibrated against gravimetric readings, however this was not necessary since it was the relative values rather absolute values that were the subject of our analyses.

2.3 Sampling sites and measurement procedures

Two sets of instruments were used to measure PNSD, PN and $PM_{2.5}$ concentrations. One measured continuously at the highest level (usually on the rooftop), which was designated as the reference site for each building. The second set measured simultaneously at one of the lower levels. The air sampled from outdoors (i.e. outside the plant room) was delivered to the instruments via a 1 m long conductive tubing, with an inner diameter of 6 mm. The locations of all outdoor air sampling points were carefully considered to avoid the influence of nearby

exhaust air from the HVAC system, if any. A flow splitter was used in cases where several instruments sampled air from the same location. Measurements were performed continuously for at least 24 hours and under different wind conditions at each of the lower level sites. The measurement campaign at each building ranged from two to three weeks. The specific measurement procedures for each of the three buildings are described below.

Building A: One set of instruments continuously measured at the reference site located on the top level (level 3) 14.5 m above the ground, 8.5 m above and 7 m away from the busway. The second set was rotated between the ground floor, level 1 and level 2 at the front of the building (facing the busway), at heights of ~1.5, 6.5 and 10.5 m above ground, respectively (see Fig. 2). The measurements were performed from the 22 July to the 16 August 2009, during the Australian winter period.

Building B: The reference site was located on the rooftop, about 78.5 m above road level, and one set of instruments sampled continuously at this location. The second set simultaneously sampled at 1.5 m above and ~ 5 m from the roadway, as shown in Fig. 3, since there were no other access points available at other levels due to the tight glass wall structure of the building. Measurements were performed from the 14 to the 30 January 2010, during the Australian summer period.

Building C: One set of instruments sampled continuously at the reference site, which was located 21.5 m above the ground, and 13.5 m above and 7 m away from the freeway. The second set was moved between sites located at heights of ~1.5 m, 5.5 m, 9.5 m and 21.5 m (levels 1, 2, 3 and 6, respectively) on the opposite side of the building to the reference site (the rear of the building). The sampling sites and building layout are shown in Fig. 4. Measurements were performed from the 24 June to the 16 July 2010, during the Australian winter period.

2.4 Meteorological data

Meteorological parameters, including wind speed, wind direction, temperature and relative humidity corresponding to each measurement campaign were obtained from the Queensland Bureau of Meteorology weather station located in Brisbane CBD between 1 to 3 km east to south east of the measurement sites. Global solar radiation was collected at the Queensland Department of Environment and Resource Management site, about 10 to 12 km south of the measurement sites. A summary of the meteorological data is provided in Table 1.

2.5 Identification of nucleation event

Morawska et al. (2008) has shown that motor vehicle emissions are the major source of air pollution in urban environments. Particles from vehicle emissions are classified as either primary or secondary. The primary particles are generated directly from engines and range in size from 30 – 500 nm. The secondary particles are formed via nucleation in the atmosphere after emissions from the tailpipe and are generally below 30 nm.

In order to identify nucleation events, contour plots of data based on a 24-hour period, from 0:00 – 24:00, were visually analysed. Criteria proposed by Dal Maso et al. (2005) and Hussein et al. (2008) were then applied to identify nucleation events. These criteria are: (i) a distinctly new mode of particles must appear in the size distribution; (ii) the mode starts in size range of < 30 nm; (iii) the mode prevails over a time period of hours; and (iv) the new mode shows signs of growth. In urban environments, nucleation events have been observed both with and without particle growth (Cheung et al., 2011; Gao et al., 2009; Park et al., 2008). Therefore, an event where the nucleation mode particle number concentrations increased during the day, but the particles did not grow larger during the event period, as indicated by a near constant Geometric Mean Diameter (GMD) value, was also considered as a nucleation event. Atmospheric conditions during the events were also recorded to identify the preconditions for nucleation process.

2.6 Data analyses

In order to compare PN concentrations in different size ranges at street and rooftop levels, PN concentrations were classified into the following size ranges: 8.5 – 30 nm, 30 – 50 nm, 50 – 100 nm, 30 – 100 nm, 100 – 300 nm and 30 – 300 nm. The number of particles within each range was referred to as $N_{<30}$, N_{30-50}, N_{50-100}, N_{30-100}, $N_{100-300}$ and N_{30-300}, respectively.

Vertical profiles of PNSD and $PM_{2.5}$ concentrations for each building were determined by normalising measured concentrations to the reference site. These were calculated as the ratio of concentrations measured at the different levels to the corresponding concentration at the reference site. Following this, the mean ratios of normalised concentrations were shifted so that the lowest height of each building was 1.0. This allowed trends of increasing or decreasing concentrations to be interpreted as values larger or smaller than one.

Statistical analyses included the Student's t-test to assess differences in mean particle concentrations between different heights and time periods. Paired PNSD and $PM_{2.5}$ concentrations corresponding to different heights at each building were analysed using the linear correlations. The 5% level was taken to indicate statistical significance in all cases.

3 Results and discussion

3.1 Variation of particle number size distribution at rooftop and street levels

Whilst 'rooftop level' refers to the reference site at each building, the 'street level' varied for each building depending on the height of the busy road close by. For example, the height of level 1 at Building A is approximately the same height as the nearby busway, and therefore, the measurements conducted at level 1 are considered to be 'street level' measurements. Similarly, the ground floor of Building B (close to city street level) and level 3 of Building C (close to the freeway) are also referred to as 'street level'.

To interpret the daily pattern of PNSD at rooftop and street levels of each building, PNSD spectra and average daily PN concentrations for $N_{<30}$, N_{30-50}, N_{50-100}, and $N_{100-300}$ were plotted

against time of the day for Buildings A, B and C (see Figs. 5, S1 and S2, respectively). In general, PNSD trends at rooftop and street levels were similar at each building.

At the rooftop and street levels of Building A, PN size fraction concentrations increased in the early morning and late afternoon. However, the concentrations in the morning were higher than those in the afternoon. During the middle of the day (noon) and early afternoon, $N_{<30}$ repeatedly increased while other particle size concentrations remained constant or decreased. At Building B, $N_{<30}$ increased significantly during the early afternoon, while other particle size range concentrations decreased at both the rooftop and street levels. Similar to Building A, all particle size concentrations at Building C increased in the early morning and late afternoon, while only $N_{<30}$ increased again around noon.

Daily mean variations of PN size fraction concentrations increased in the early morning and late afternoon at Buildings A and C. Traffic flows on the streets close to the sampling sites also showed corresponding peaks during these times, which indicate the influence of vehicle emissions on increased particle concentrations during the rush hours. In contrast, $N_{<30}$ concentration increased at noon, while other particle size ranges remained constant or decreased at both the rooftop and street levels of all three buildings. In addition, the traffic flow rates decreased around midday. This could suggest the occurrence of new particle formation during this period. A detailed analysis and discussion of the influence of vehicle emissions and new particle formation on particle concentrations is provided in the following section.

3.2 Influence of vehicle emissions and new particle formation on PNSD and $PM_{2.5}$ concentrations at rooftop and street levels

3.2.1 Influence of vehicle emissions on PN and $PM_{2.5}$ concentrations at rooftop and street levels

The days that did not meet at least one of the criteria for the nucleation event definition were defined as a non- or unclear nucleation event day. Based on this, there were 19, 8, and 20 days

that were classified as non- or unclear nucleation event at Building A, B, and C, respectively. Weekdays characterised by non- or unclear nucleation events were selected to assess the influence of vehicle emissions on the PN and $PM_{2.5}$ concentrations at the rooftop and street levels of each building. Examples of PNSD spectra, PN and $PM_{2.5}$ time series plots at the rooftop and street levels of Buildings A, B and C, as well as their ratios are presented in Figs. 6, 7, S3, S4, S5 and S6, respectively. Statistical results are given in Table 2.

From Fig. 7 it can be seen that both PN and $PM_{2.5}$ concentrations peaked at the rooftop and street levels of Building A during the early morning on the 7 August 2009. However, PN concentration at the rooftop level was significantly higher than at street level, while the opposite was the case for $PM_{2.5}$. The bus ramp located close to Building A may explain the higher PN and $PM_{2.5}$ concentrations in the morning rush hours compared to those in the afternoon rush hours. About 75% (157/209) of buses during the morning rush hour have to ascend an uphill ramp, and these would have greater emissions than those during the afternoon rush hours that predominantly travel downhill.

PN concentration at the rooftop and street levels of Building B on the 18 January 2010, fluctuated according to the wind conditions during the day. However, both PN and $PM_{2.5}$ concentrations at street level were significantly higher than those at the rooftop level during the morning and afternoon rush hours when the wind blew from SW and NE directions. This can be explained by the one-way city street immediately adjacent to the lower sampling site at Building B, which had a traffic flow from the SW to the NE and therefore both SW and NE winds blew parallel the street. Given that the NE wind blew against the traffic flow, it was classified as up-canyon wind, while the SW wind was classified as down-canyon wind. Both PN and $PM_{2.5}$ concentrations at the rooftop and street levels were significantly higher during up-canyon wind (in the afternoon) compared to down-canyon wind (in the morning) (refer to Table 2 for comparative results) and ratios between the street and rooftop levels for both PN

and $PM_{2.5}$ concentrations were also significantly higher during the up-canyon wind compared to the down-canyon wind.

At Building C, PN and $PM_{2.5}$ concentrations at the roof top level were significantly higher than those at street level during the morning rush hours on the 6 July 2010. Low dispersion due to low wind speed ($v = 0.31 \pm 0.29$ m s^{-1}) during this time might explain why the particle concentrations at the rooftop sampling point, which was closer to the freeway, were higher than those at the opposite sampling point at street level. During the afternoon, a WNW wind blew almost parallel to the freeway and the building, resulting in a better dispersion of pollutants on both sides of the building and also being the likely explanation why the PN and $PM_{2.5}$ concentrations were not significantly different at the rooftop and street levels (*p*-values of 0.06 and 0.45, respectively).

In summary, time series of PN and $PM_{2.5}$ concentrations and their ratios between the rooftop and street levels showed clear diurnal variation. As expected, vehicle emissions strongly influenced both PN and $PM_{2.5}$ concentrations at both levels, especially during the rush hours at all three buildings. Similarly, building topography, distance to the emission sources, and wind speed and direction also had an observed effect on particle concentrations at the 3 buildings.

3.2.2 Influence of new particle formation on PNSD and $PM_{2.5}$ concentration at rooftop and street levels

Based on the inclusion criteria for nucleation identification, we observed 7 events during a 3 weeks measurement campaign at Building A, 9 events during a 2 weeks measurement campaign at Building B and 3 events during a 3 weeks measurement campaign at Building C. The frequency of nucleation events at Building B (measured during summer) was clearly higher than those at Buildings A and C (measured during winter), which is in agreement with the findings of Qian et al. (2007) and Mejia and Morawska (2009). A summary of the conditions observed during the nucleation events is provided in the Supplementary Table S1.

Representative nucleation events were selected to analyse the influence of new particle formation on PNSD at the rooftop and street levels of each building, to assess their likely sources and impact on vertical profiles. PNSD spectra, time series' of $N_{<30}$, $N_{30\text{-}100}$ and $PM_{2.5}$ concentrations, as well as ratios of PN and $PM_{2.5}$ concentrations at the rooftop and street levels of Buildings A, B and C are presented in Figs. 8, 9, S7, S8, S9 and S10, respectively. The results of statistical tests are presented in Table 3.

$N_{<30}/N_{30\text{-}300}$, which is the ratio between nucleation mode and accumulation mode PN concentration, was used by Kumar et al. (2009) to evaluate the rate of production of new nucleation mode particles. When analysed together with $N_{<30}$, which indicates nucleation mode PN concentration, it is possible to assess the strength of new particle formation at the different levels of each building. From Table 3, it can be seen that both $N_{<30}$ and $N_{<30}/N_{30\text{-}300}$ were significantly higher at the rooftop level compared to street level at each building, and they were also clearly higher at Building B than at Buildings A and C. Meanwhile the rooftop $PM_{2.5}$ concentration was significantly lower than the street level $PM_{2.5}$ at all three buildings.

Based on the higher values of $N_{<30}$ and $N_{<30}/N_{30\text{-}300}$ at the rooftop level of each building, we inferred that the production of new nucleation mode particles was stronger at the rooftop level than the street level at all three buildings. Vakeva et al. (1999) reported two important factors that can favour a much greater production of particles by local vehicle emissions: (i) a higher concentration of condensable gases, and (ii) a smaller concentration of pre-existing particles. Additionally, both O Dowd et al. (1999) and Boy and Kulmala (2002) identified the important role of solar radiation on new particle formation. The roles of these factors in initiating the events we observed are discussed below.

Wind direction during the nucleation event at Building A on the 3 August 2009, was WNW. In this case, both sampling sites and the busway were on the downwind side of the building. Leuzzi and Monti (1998) modelled the dispersion of a tracer gas emitted from a line source located downwind of a building and reported that high pollutant concentrations occurred at

locations corresponding to the vortex on the leeward side of the building. At about 40 m wide and 17 m high, Building A can be considered a wide and low building and therefore the vortex, which entrains the smaller particles or condensable gases emitted from vehicles, probably formed at a level higher than the street level, while the larger or pre-existing particles (mainly attributed to $PM_{2.5}$) remained suspended and stagnated at the lower levels. Therefore, it appears that the stronger nucleation observed at the rooftop compared to the street level was due to higher condensable gas and lower pre-existing particle concentrations.

Leuzzi and Monti (1998) also modelled an upwind line source and reported that low concentrations occurred on the leeward side of the building, with only a small amount of pollutants able to penetrate into the region. During the nucleation event at Building C on the 8 July 2010, a SSW wind blew perpendicular to the building from direction of the freeway. Therefore, the rooftop sampling site was upwind and received pollutants directly from the freeway emission sources, while the street level sampling site was located in the lee of the building. This suggests that there were lower concentrations of condensable gases at the street level compared to the rooftop level of Building C and that the higher $PM_{2.5}$ concentrations measured at street level might be due to the stagnation of larger, pre-existing particles on the leeward side of the building.

Based on $N_{<30}$ and $N_{<30}/N_{30-300}$ at rooftop and street levels, we also concluded that the intensity of new particle formation at Building B on the 16 January 2010, was clearly stronger than that at Buildings A and C, although the mean solar radiation intensity (W m^{-2}) (Mean ± 95% CI) during the nucleation event at Building B was not significantly different compared to Building A (664.3 ± 20.7 vs. 689.4 ± 22.4, $p = 0.36$). At the same time, ratios between rooftop and street level values for $N_{<30}$ and $N_{<30}/N_{30-300}$ were significantly lower at Building B compared to those at Building A (1.15 ± 0.09 vs. 1.88 ± 0.27, $p < 0.01$; 1.20 ± 0.14 vs. 1.84 ± 0.30, $p < 0.01$, respectively). The nucleation event observed at Building B occurred on a weekend, when vehicle density was typically low and a strong NE wind (3.57 ± 0.32 m s^{-1})

was blowing. The resultant increase in $N_{<30}$ but decrease in N_{30-100} suggests that the PN concentrations at the sampling site were not significantly influenced by local vehicle emissions but more likely from upwind air masses. In this case, the air mass was likely to come from an industrial zone about 15-18 km NE of the city. Further analysis and comparison of the data measured at this building was conducted along with data collected from a Queensland Department of Environment and Resource Management station, which is about 10 km SW of the Brisbane city and 25 km SW of the NE Brisbane industrial zone. The results showed similar trends in PN concentrations between the two locations during the NE winds, but not for other wind directions, during the nucleation days. This implies that emissions from the NE Brisbane industrial zone are those which contribute to the PN concentrations in the Brisbane CBD and surrounding areas. Furthermore, a similar phenomenon was identified and reported by Cheung et al. (2011) in the Brisbane region. It should also be noted that newly formed particles at both the rooftop and street levels did not show signs of growth (their GMDs were almost constant during the event). This indicates that the newly formed particles already underwent growth before reaching the monitoring sites and they were likely to be relatively homogeneous in size when reaching Building B after the distance travelled. Furthermore, the NE wind, which would have blown parallel to the street canyon, and minimal turbulence due to the low vehicle density could explain why the difference in PN concentrations (cm^{-3}) between the rooftop and street levels at Building B (16,900 ± 1,490 vs. 15,650 ± 1,470; $p < 0.05$) was significant, but not to the same extent observed at Buildings A (8,160 ± 1,020 vs. 4,570 ± 280; $p < 0.01$) and C (5,340 ± 450 vs. 3,310 ± 270; $p < 0.01$). This new finding contradicts the results reported for Building A and locations investigated by Kumar et al. (2009), where new particle formation was mainly influenced by local vehicle emissions. This also has implications for modelling urban canyon PN concentrations for both planning and exposure assessment purposes, and indicates the value of location-specific measurements at underpinning these.

In summary, the time series concentrations of $N_{<30}$, N_{30-100} and $PM_{2.5}$, as well as the time series ratios of PN and $PM_{2.5}$ concentrations at the rooftop and street levels showed that new particle formation events influenced and contributed to increases in PN concentrations at both rooftop and street levels at all three buildings. However, the factors that contributed to the observed phenomena were different between the three buildings. At Building A and C, the new particles were mainly formed from local vehicle emissions and therefore, the formation process was expected to depend mainly on local conditions, such as high condensable gas concentrations and solar radiation intensity, together with low pre-existing particle concentrations. Meanwhile at Building B, the newly formed particles were blown in from the direction of a nearby industrial zone and therefore, new particle production was not the result of local sources but was strongly influenced by wind speed, wind direction and the origin of incoming air masses. Detailed consideration of the factors described above should be undertaken prior to modelling urban canyon particle concentrations and profiles, and a 'one-size-fits-all' approach is likely to be unable of accounting for the specific determinants at each individual building.

Nucleation events are often studied in the context of their role as physical phenomena, and typically within the context of producing natural and anthropogenic aerosols that may affect climate change. This study has shown an underappreciated role of nucleation in producing particles that can affect large numbers of people, due to the high density and occupancy of urban office buildings and the fact that the vast majority of people's time is spent indoors.

3.3 Vertical profiles of particle concentrations

The average vertical profiles of the PNSD and $PM_{2.5}$ for the entire day, rush-hours and during nucleation events at Buildings A, B, and C are presented in Figs. 10, 11 and 12, respectively. It should be noted that the data of the nucleation events at Building C were only collected at rooftop and street levels and therefore, constructing a vertical profile based on nucleation events at this building, was not appropriate. However, the measured results at Building C

show that the PN concentration at rooftop levels was significantly higher than at street levels during the event, while the opposite was the case for the $PM_{2.5}$ concentration.

At Building A, the trends of total number concentration (TNC) and $N_{<30}$ were similar. Their concentrations during nucleation events themselves and over 24 hour on the day of nucleation events constantly increased with height ($p < 0.01$). While during the rush-hours, they decreased between 1.5 and 10.5 m, and then increased onward ($p < 0.05$). In contrast, the trends of $N_{30\text{-}100}$ and $N_{>100}$ fluctuated and depended on the measurement heights and times. In general, the daily $PM_{2.5}$ concentrations decreased with increasing height, however they stabilised at heights between 6.5 and 10.5 m. During rush-hours, $PM_{2.5}$ concentrations were higher at heights of 6.5 and 10.5 m, but lower at a height of 14.5 m, compared to the daily concentrations ($p < 0.05$). The $PM_{2.5}$ concentrations during the nucleation events were generally lower than the daily concentrations ($p < 0.01$).

At Building B, $N_{30\text{-}100}$, $N_{>100}$ and $PM_{2.5}$ concentration at street levels were always higher than those at rooftop levels ($p < 0.05$). The daily and rush-hour TNCs were significantly higher at street level compared to those at rooftop level, but the opposite was the case during the nucleation events ($p < 0.05$). $N_{<30}$ at rooftop level was significantly higher than at street level during the nucleation event ($p < 0.01$), while their daily and rush-hour concentrations were relatively similar (*p*-values of 0.17 and 0.78, respectively).

The daily PNSD and $PM_{2.5}$ concentration decreased with height between 1.5 and 21.5 m at the rear (opposite side facing the road) of Building C ($p < 0.01$), however $N_{30\text{-}100}$, $N_{>100}$, $PM_{2.5}$ tended to stabilise at heights between 5.5 and 9.5 m, followed by a less pronounced decrease from 9.5 to 21.5 m. During the rush-hour periods, $N_{30\text{-}100}$, $N_{>100}$, TNC decreased from 1.5 to 9.5 m, and then stabilised at heights between 9.5 and 21.5 m. $N_{<30}$ increased at the beginning of the rush-hour period, then decreased from 5.5 to 9.5 m, and finally stabilised onwards. The rush-hour $PM_{2.5}$ followed the $PM_{2.5}$ daily trends and was higher than the daily concentrations.

In general, the trend of TNC followed those of $N_{<30}$ and N_{30-100} during the nucleation event and rush-hours, respectively, while the trends of $N_{>100}$ and $PM_{2.5}$ were similar.

At Building B, the daily and rush-hour PN concentrations at street level were higher than those on the rooftop. This finding is in agreement with the results of previous studies (Hitchins et al., 2002; Kumar et al., 2009; Li et al., 2007; Longley et al., 2004; Väkevä et al., 1999). On the contrary, the daily and rush-hour PN concentrations at Building A increased with height. This is likely to be attributed to the fact that the busway is located close to the building and elevated above ground level, and therefore, it has a stronger influence on the concentrations measured at higher levels compared to Building B. The daily and rush-hour PN concentrations at the rear of Building C decreased with increasing height. This finding is not in agreement with the results reported by Hitchins et al. (2002) based on measurements in Brisbane, where a short time measurement (5 samples during 450 seconds for each level) was conducted. The difference could be due to the highly diurnal variations of influencing factors, such as vehicle emissions, wind speed and wind direction on particle concentrations between the different levels of this building.

The $PM_{2.5}$ concentrations seemed to consistently decrease with height throughout the day and this finding is also in accordance with previous research (Chan and Kwok, 2000; Horvath et al., 1988; Micallef and Colls, 1998; Rubino et al., 1998). However, the $PM_{2.5}$ concentrations at Buildings A and C did not decrease consistently. In the case of the Building A, this may be due to the influence of the proximity of the busway. The sampling points were located on the rear side of Building C and were obstructed by other buildings located behind it, and therefore, some stagnation of air in this region may have influenced the $PM_{2.5}$ concentrations at mid-height levels.

In general, the vertical profiles of the $PM_{2.5}$ concentrations around the building envelopes decreased with increasing height. However, vertical profiles of the PNSD were building-specific and the rate of change with height was different at all three buildings. The results

indicate that it is not only vehicle emissions that influence the particle vertical profiles, but new particle formation as well; while particle number increased, we observed a reduction in particle mass during the nucleation events. These results serve to further define the specific effect of roadway proximity and nucleation formation on the vertical profiles of PN and $PM_{2.5}$ concentrations around building envelopes. Moreover, the highly building-specific nature of the profiles and factors affecting them underscores that, ideally, measurements form the basis of any modelling or planning exercise prior to or after construction of a building. Such an approach, which is currently lacking for the most part, will ensure the greatest model veracity. This has important implications for selecting appropriate sites for the air intakes of building HVAC systems to minimise occupant exposure to combustion products, and also to investigate how street-level exposures may be mitigated via improved design practices.

3.4 Relationship between PNSD and $PM_{2.5}$ concentration

Spearman's correlation coefficients (rho) for the PNSD and $PM_{2.5}$ concentrations at different heights and different time periods at Buildings A, B and C are presented in Figs. 13, 14, 15, respectively, and Table S2. However, as noted, new particle formation data was collected only at the reference site and street level during the measurement campaign of Building C. Therefore, correlations between the PNSD and $PM_{2.5}$ during the nucleation events at this site were not calculated. In general, the correlation coefficients between $N_{>100}$ and $PM_{2.5}$ were higher, while the correlation coefficients of $N_{<30}$ were usually lower compared to other particle size fractions.

The PNSD and $PM_{2.5}$ correlation coefficients on the rooftop were higher than those at street level at Building B. The difference between correlation coefficients for PN size fractions and $PM_{2.5}$ concentrations at Building A were higher than at Building B. This is likely due to the relative proximity of the particle sources at each level, as well as to the closeness to the busway at Building A. Both daily and rush-hour correlation coefficients of PNSD at the rear

of Building C initially increased from the ground to level 3, and then decreased closer to the rooftop.

Correlations between the PNSD and $PM_{2.5}$ were characterised by a significant variability and dependence on particle size fraction, measured height and particle emission sources. The linear correlations for the building envelopes, especially during the rush-hour and nucleation events, fluctuated significantly. This indicates that it is not appropriate to use particle mass concentrations to infer PN concentrations when modelling vertical concentrations around the building envelope and at a street level. This finding, while not a novel observation, adds weight to the existing case for separately considering particle mass and number during any urban modelling or exposure assessment exercise.

4 Conclusions

In general, vertical profiles of $PM_{2.5}$ concentrations around building envelopes showed a consistent decrease in concentration with increasing distance from nearby streets. However, vertical profiles of PN size fraction concentrations were building-specific and its rate of change was inconsistent with height. These results are not unexpected, in view of the complex flow patterns around the building envelopes, as well as in the busway and street canyons proximate to some of the buildings. The results of simultaneous measurements indicated that it was not only vehicle emissions but new particle formation was also found to strongly influence the vertical profiles of particle concentrations. Time series ratios of PN and $PM_{2.5}$ concentrations at street and rooftop levels showed clearly diurnal variation. These suggest that it is impossible to generalise vertical profiles of particle concentrations for all buildings, and that there is a need to conduct measurements or model these vertical profiles for a specific case when planning building morphology and air intake locations. Furthermore, newly formed particles and building-scale variability should also be into account when modelling particle concentrations around the building envelope, and also for urban environments and the exposures that occur within them in general.

The results of this serve to provide better insight into the impact of nucleation and local scale variability on particle concentrations, and will also help to better define particle behaviour and variability around building envelopes, which has implications for studies of both human exposure and particle dynamics.

Acknowledgement

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FIGURES AND TABLES

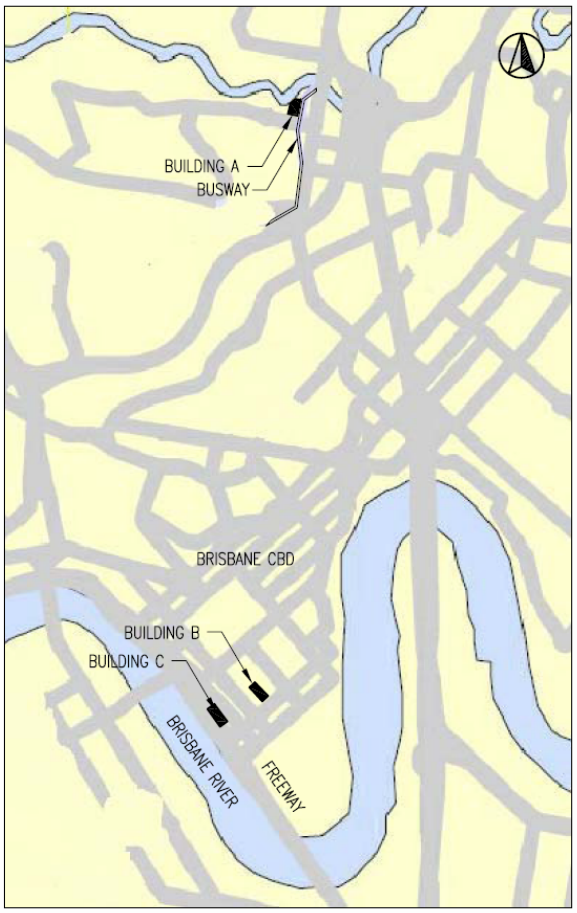


Fig. 1. Locations of Buildings A, B, and C in Brisbane.

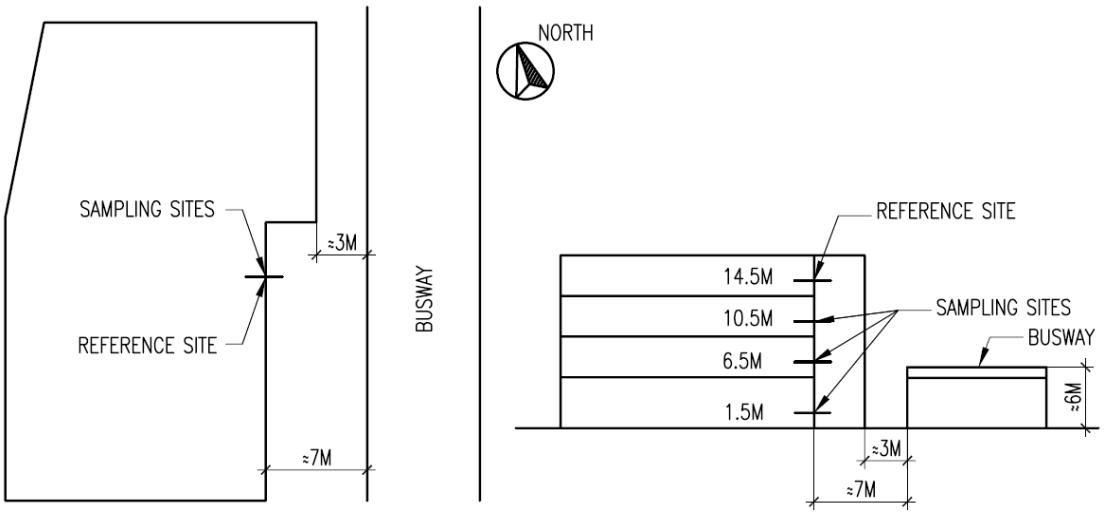


Fig. 2. Schematic diagram of Building A showing the location of the sampling points.

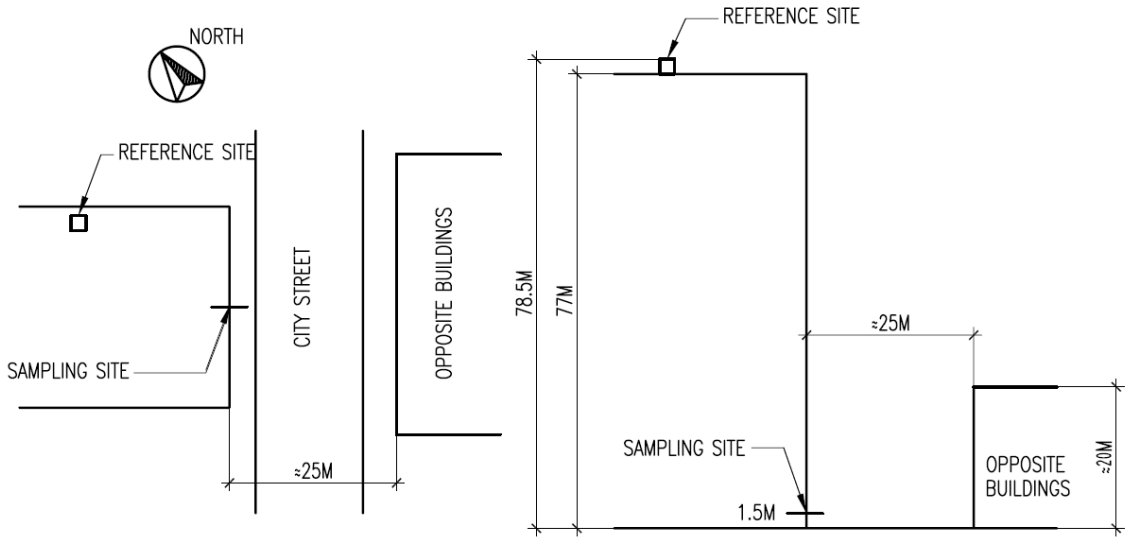


Fig. 3. Schematic diagram of Building B and the location of the sampling points.

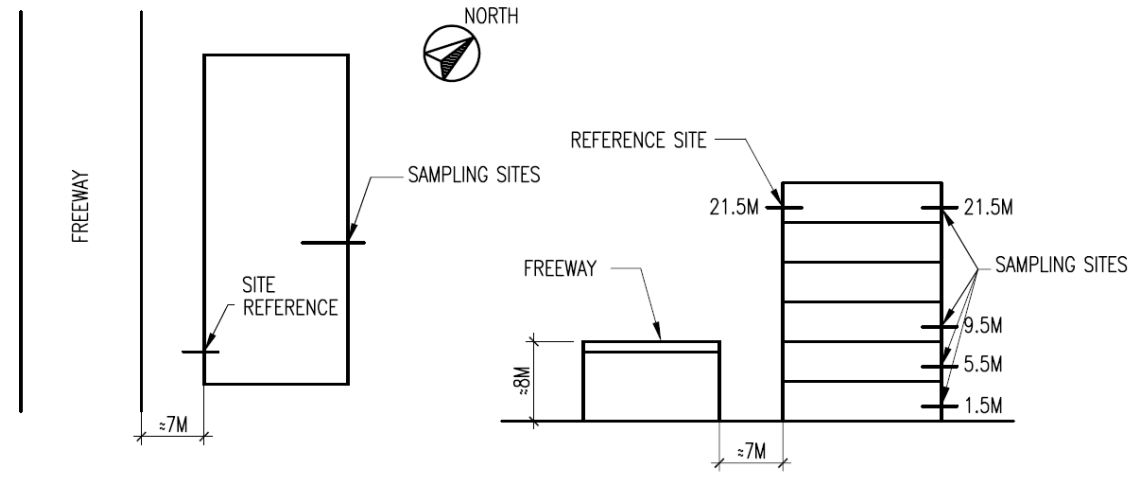


Fig. 4. Schematic diagram of Building C showing the location of sampling points.

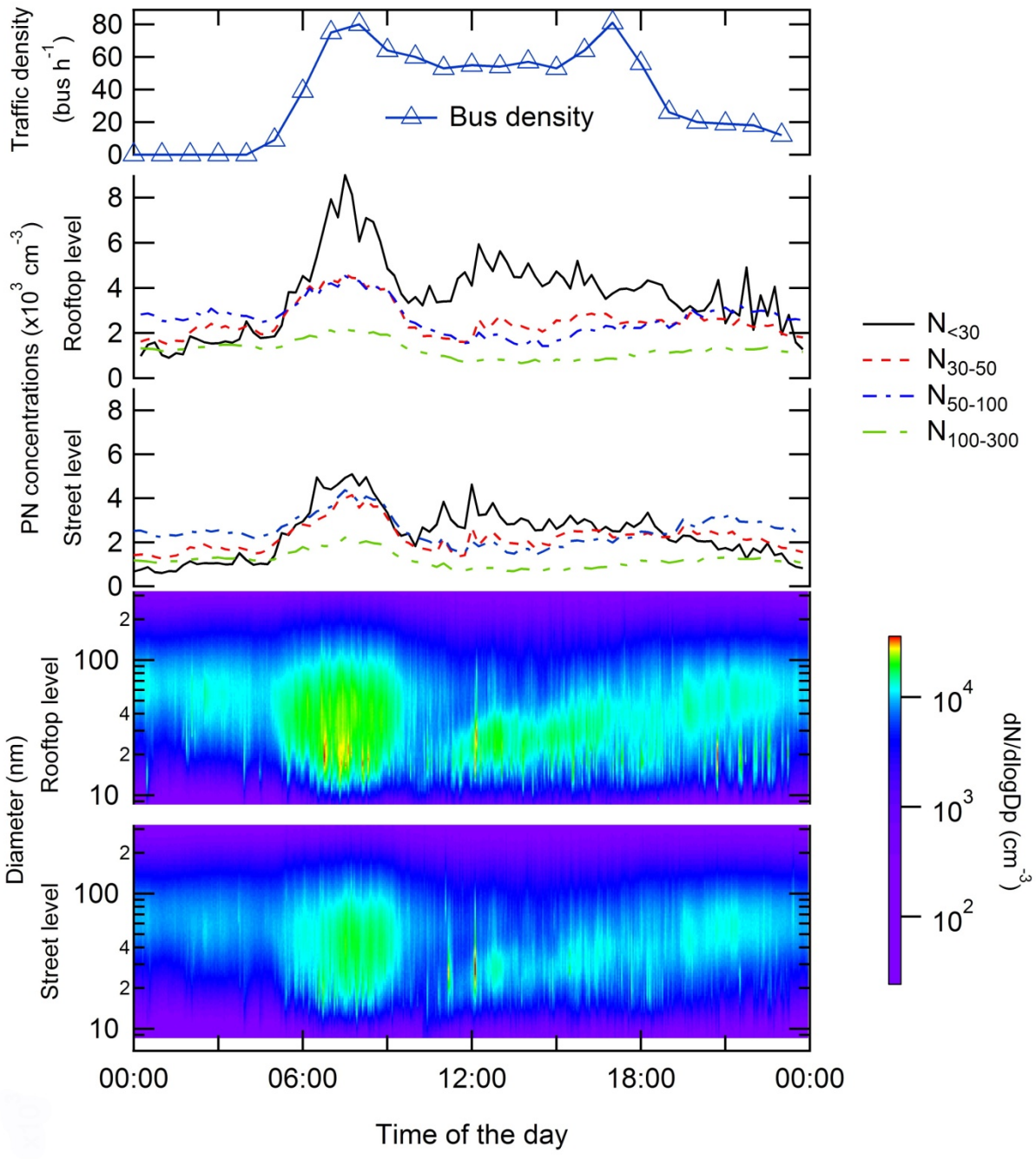


Fig. 5. Daily variation in PNSD and PN size fraction concentrations at Building A.

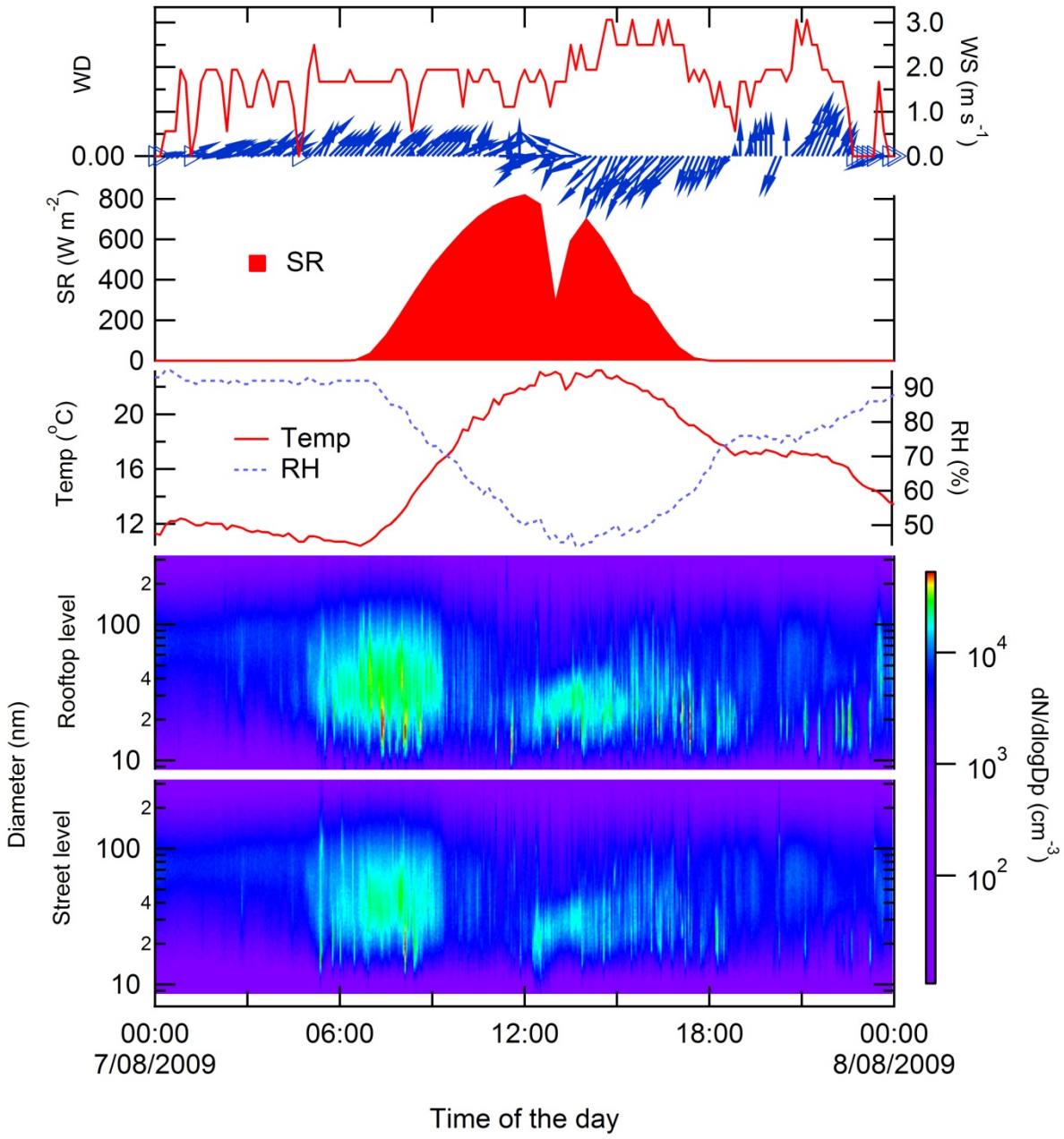


Fig. 6. PNSD spectra at Building A on a week day characterised by the non- or unclear nucleation events.

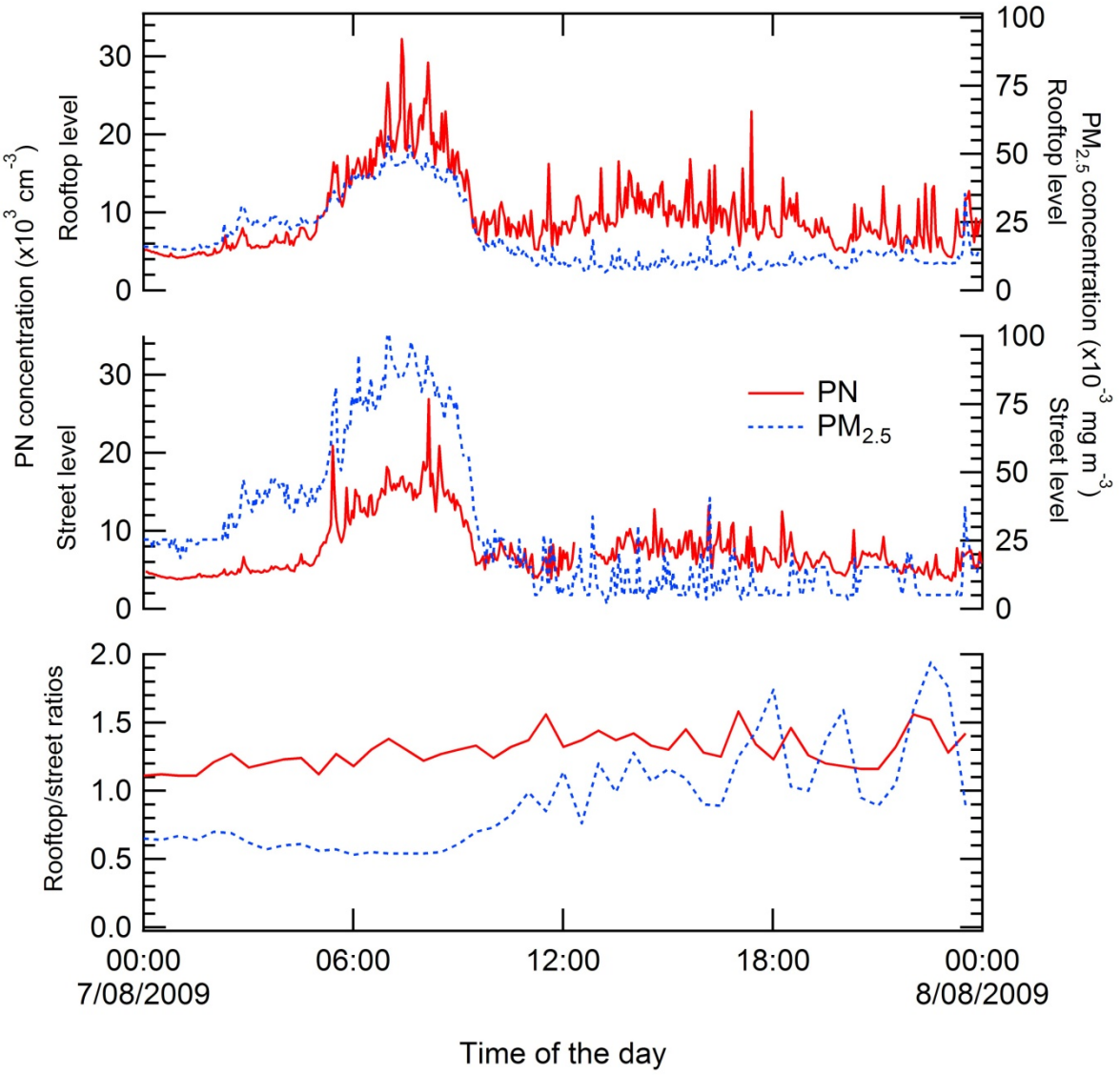


Fig. 7. Average particle concentrations and their rooftop to street level ratios at Building A on a week day characterised by the non- or unclear nucleation events.

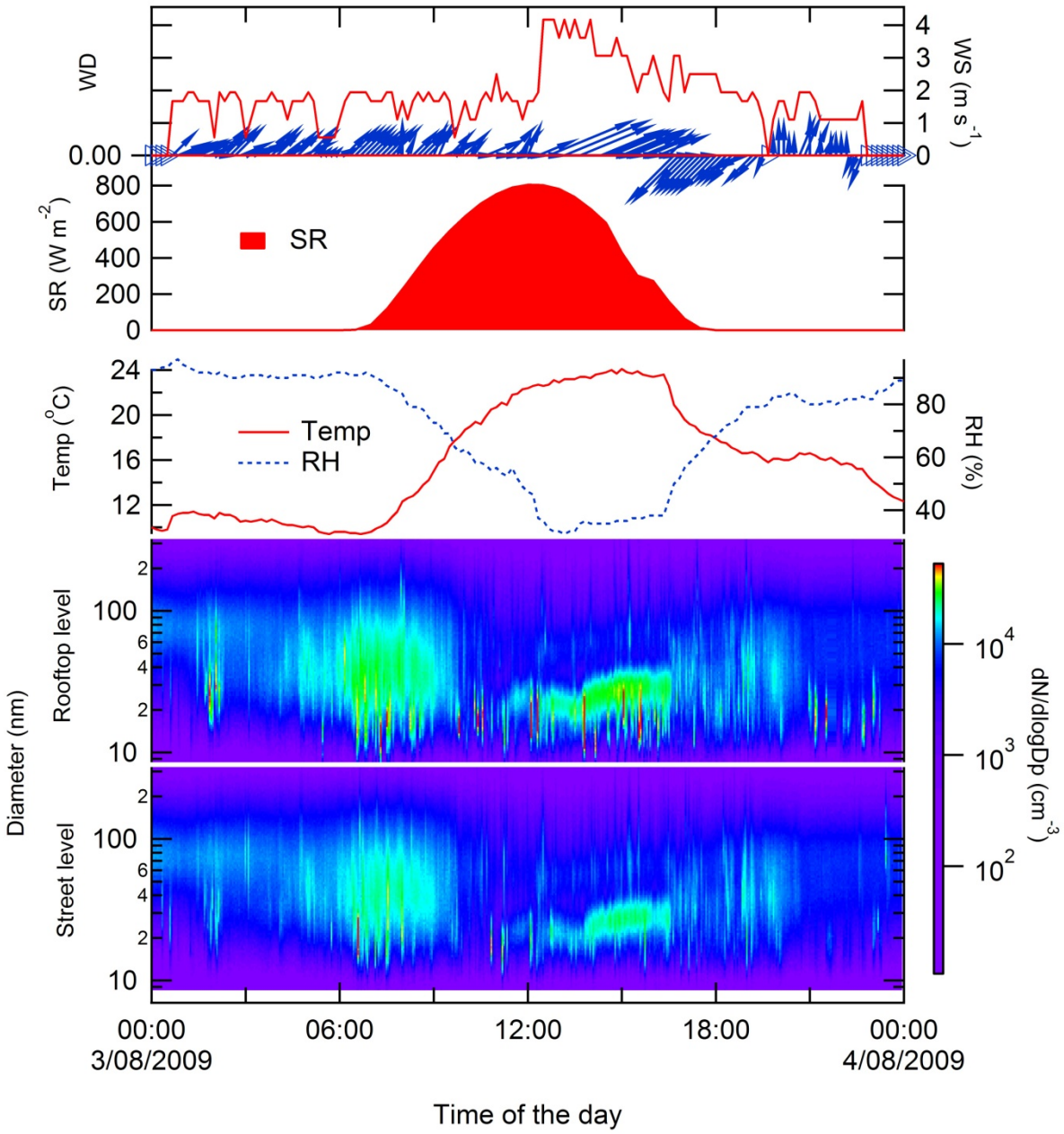


Fig. 8. PNSD spectra at Building A on a nucleation event day.

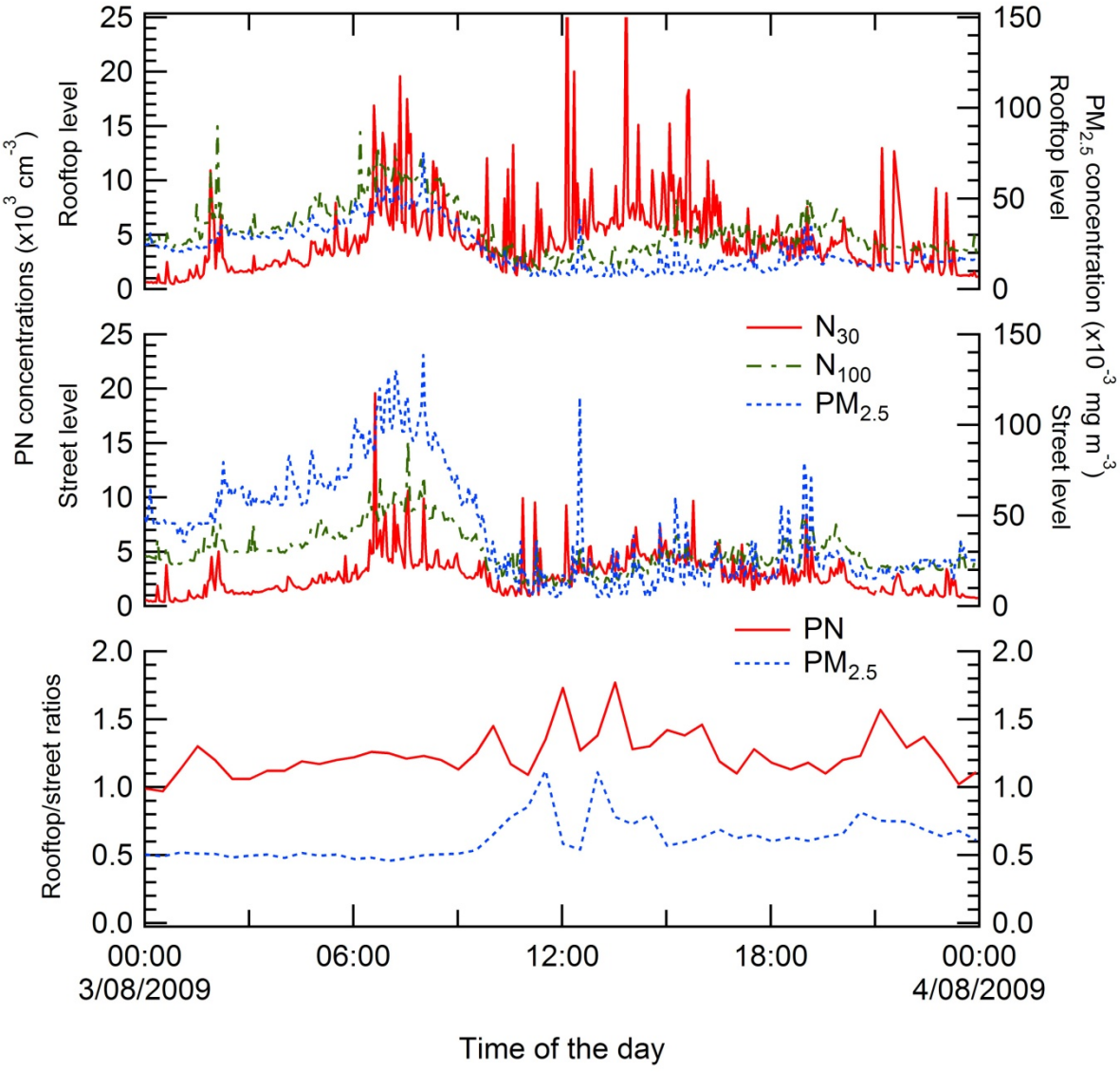


Fig. 9. Particle concentrations and their rooftop to street level ratios at Building A during a nucleation event day.

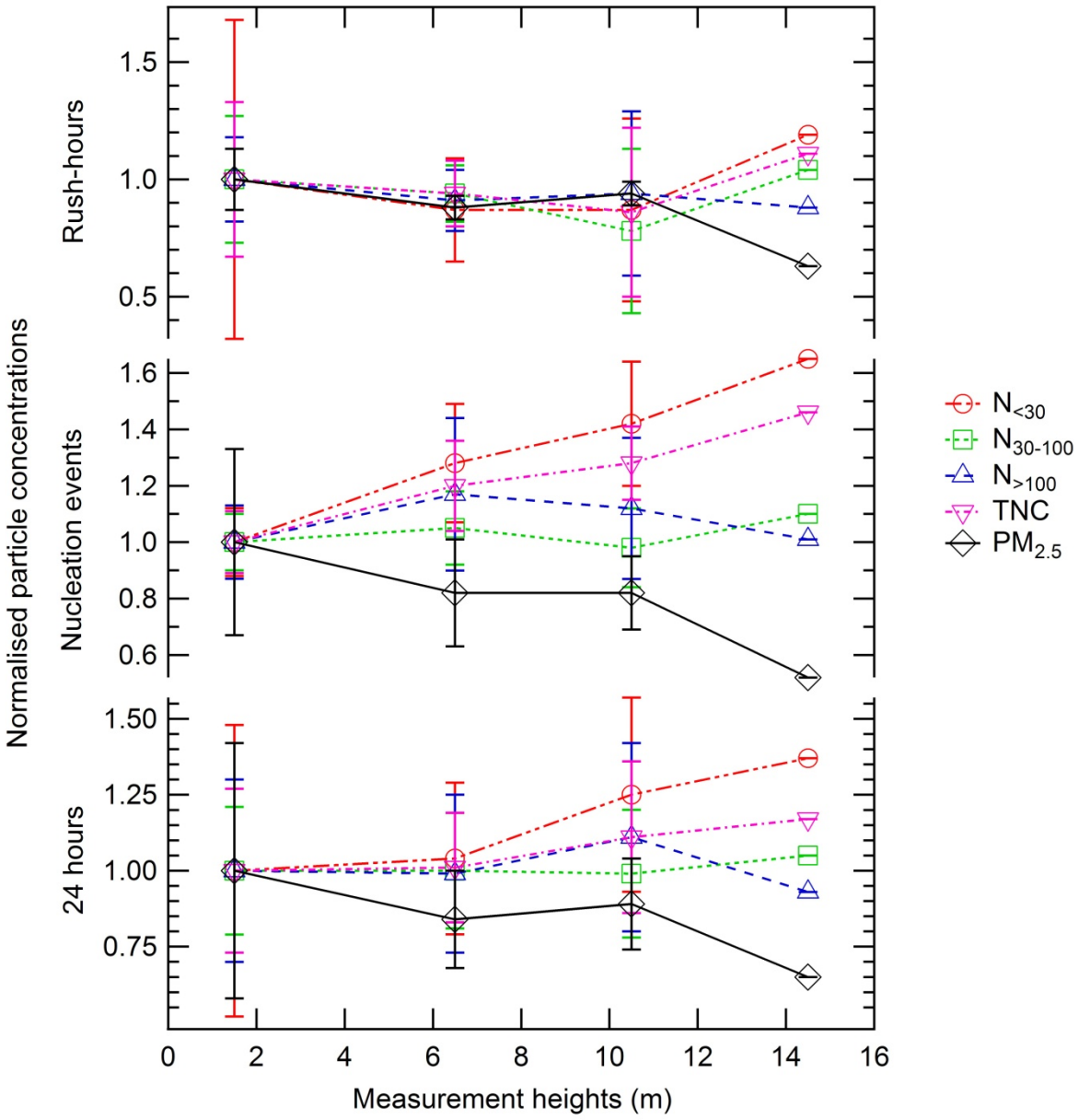


Fig. 10. Vertical profiles of PNSD and $PM_{2.5}$ concentration around Building A. Error bars denote one standard deviation.

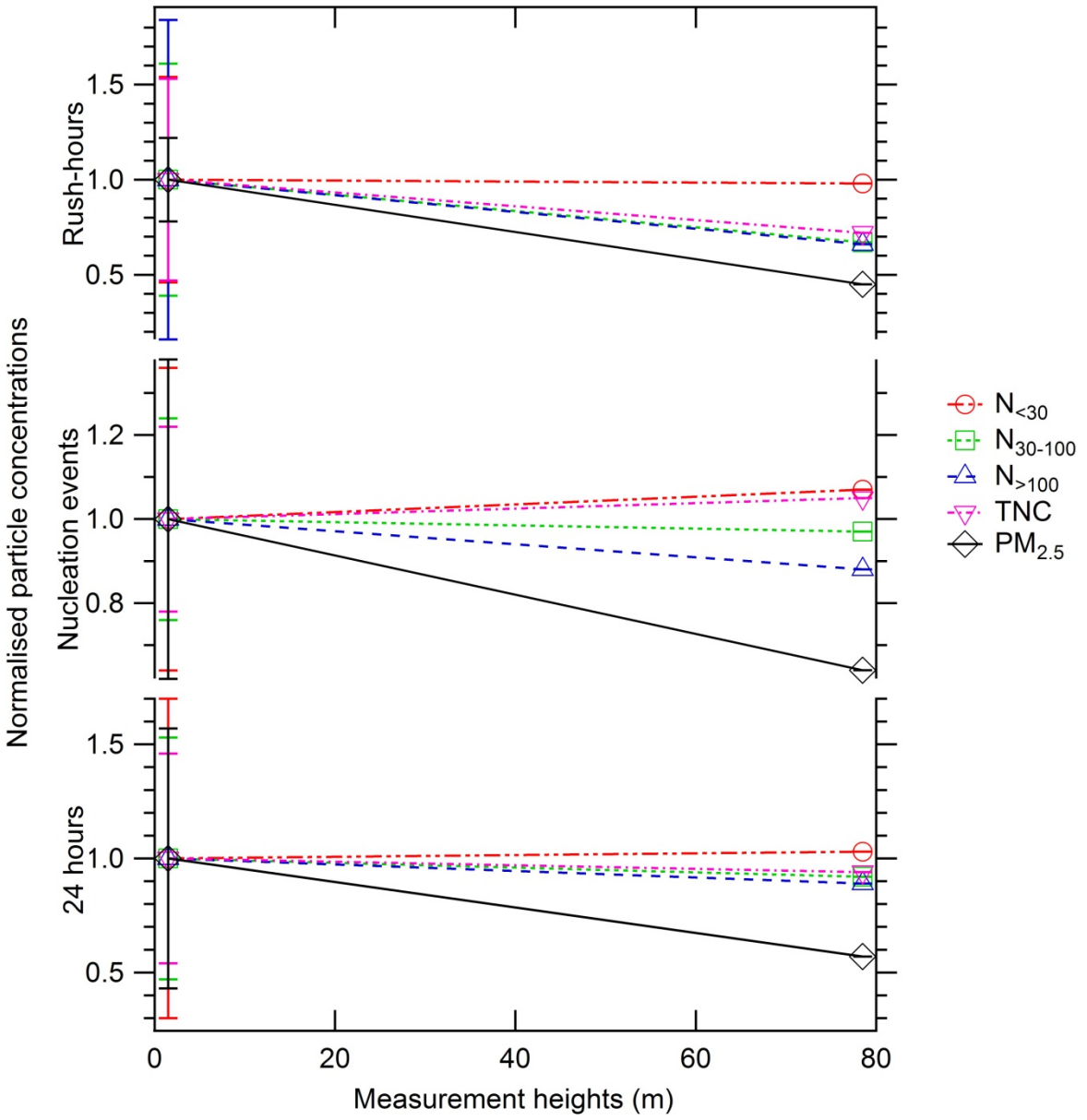


Fig. 11. Vertical profiles of PNSD and $PM_{2.5}$ concentration around Building B. Error bars denote one standard deviation.

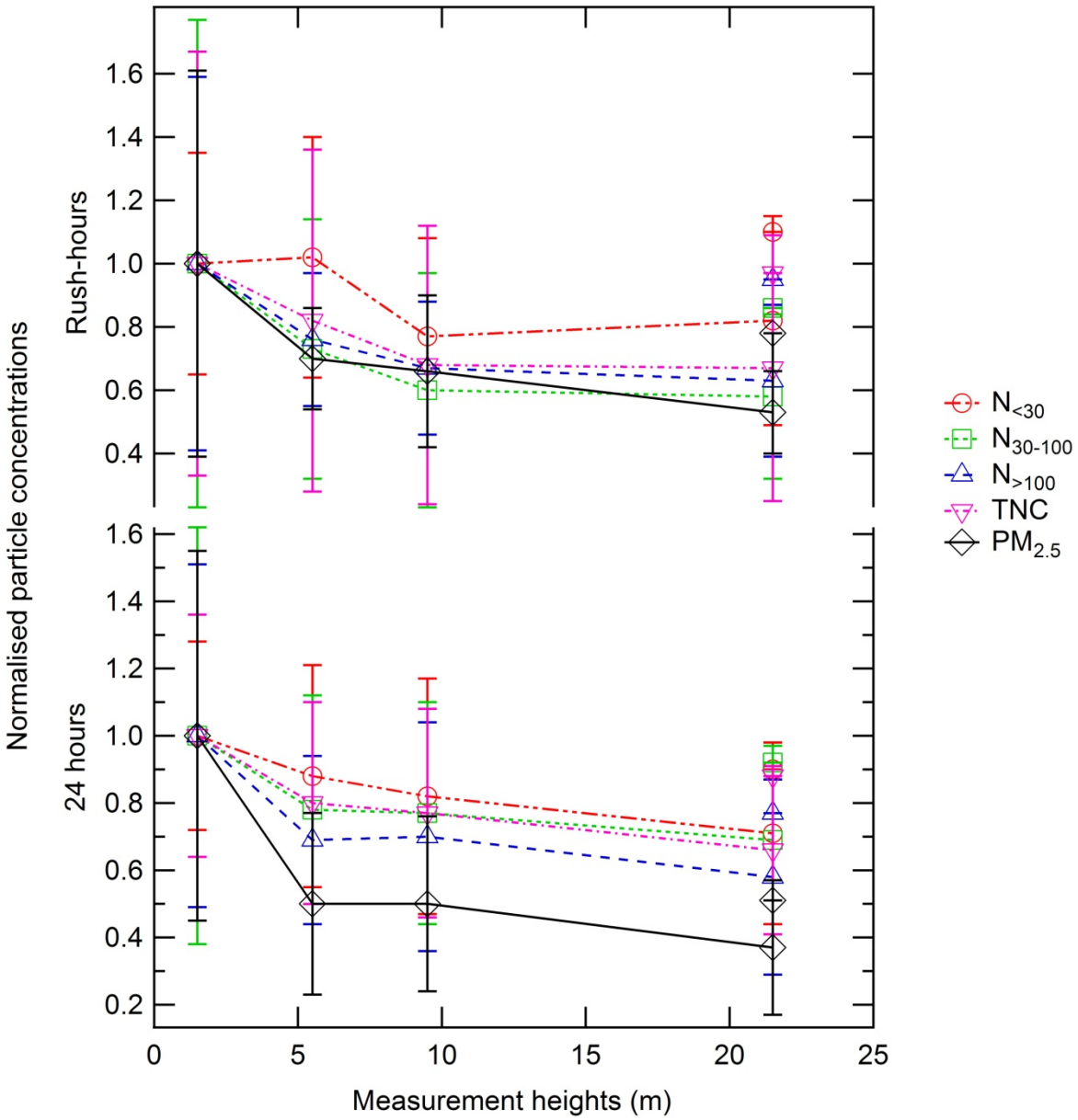


Fig. 12. Vertical profiles of PNSD and $PM_{2.5}$ concentration around Building C. Error bars denote one standard deviation.

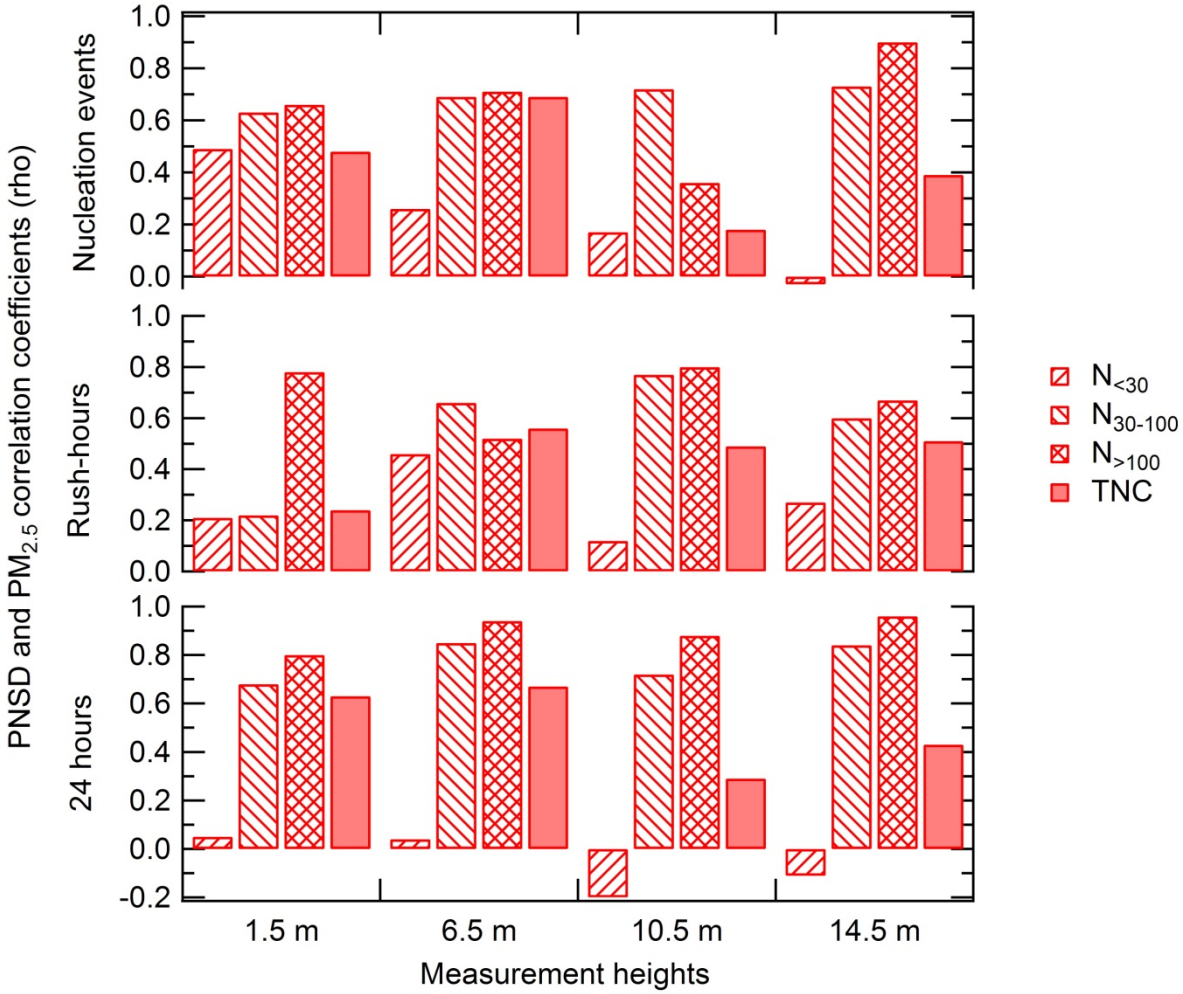


Fig. 13. Relationship between PNSD and $PM_{2.5}$ at different heights for Building A.

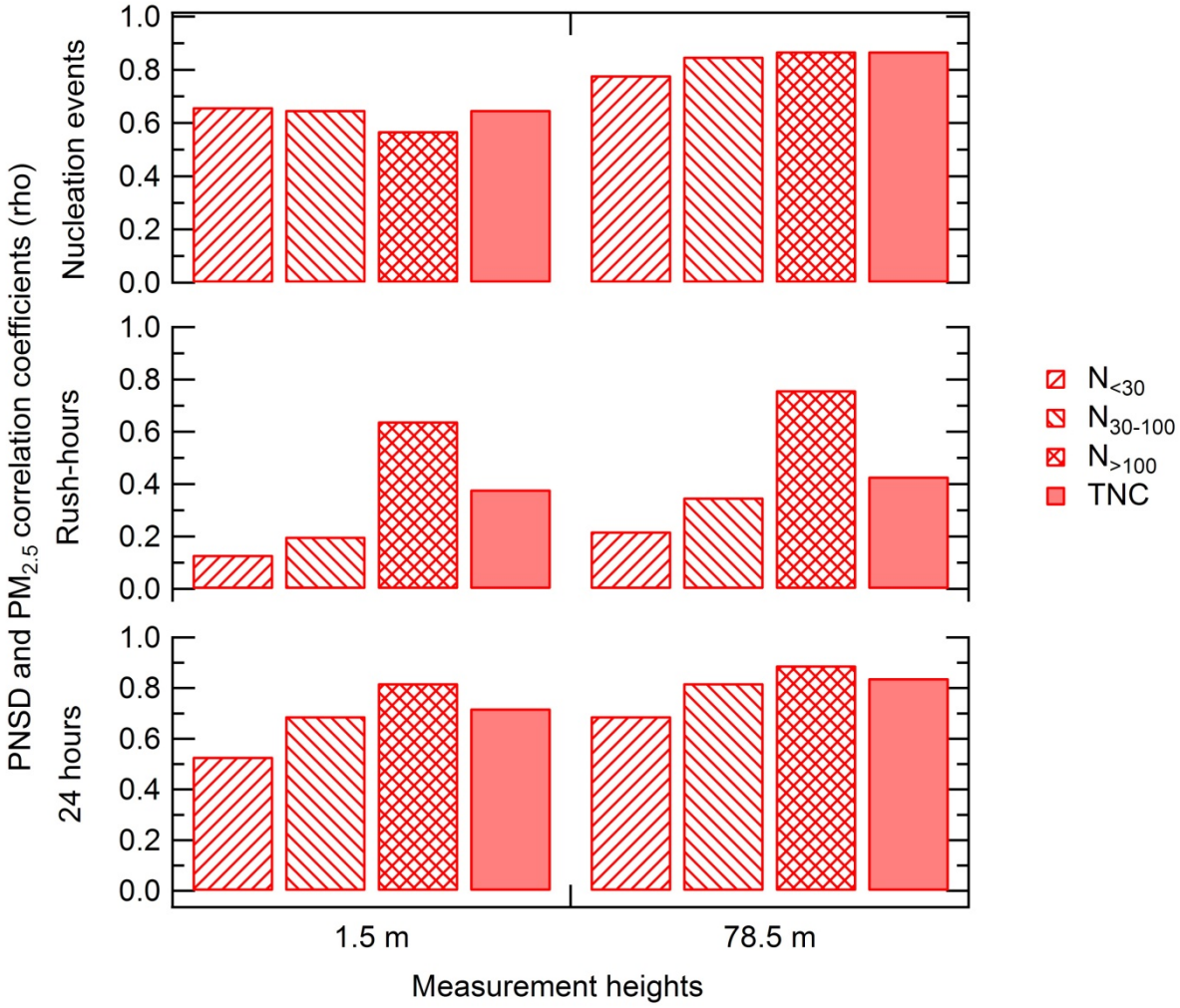


Fig. 14. Relationship between PNSD and $PM_{2.5}$ at different heights for Building B.

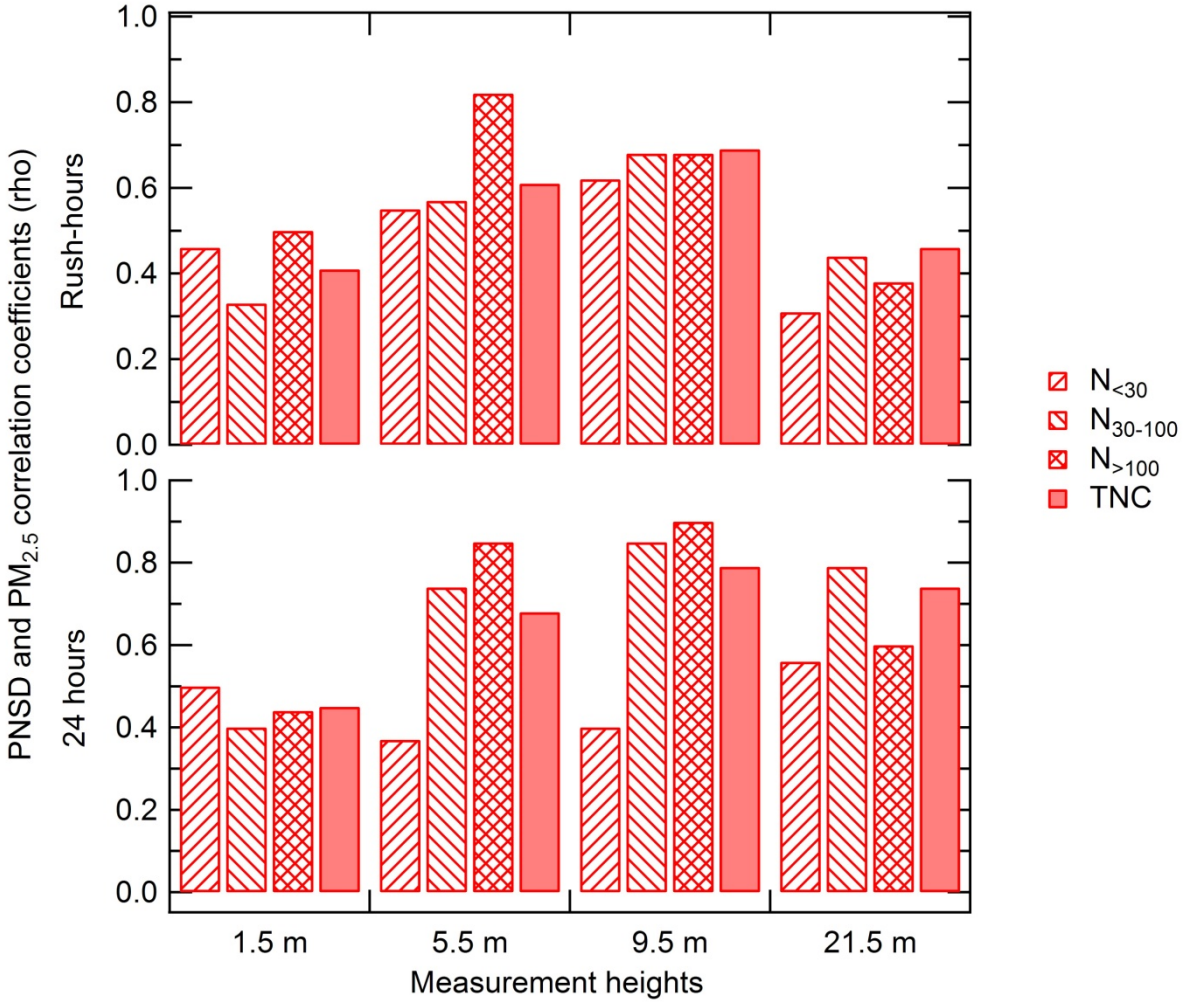


Fig. 15. Relationship between PNSD and $PM_{2.5}$ at different heights for Building C.

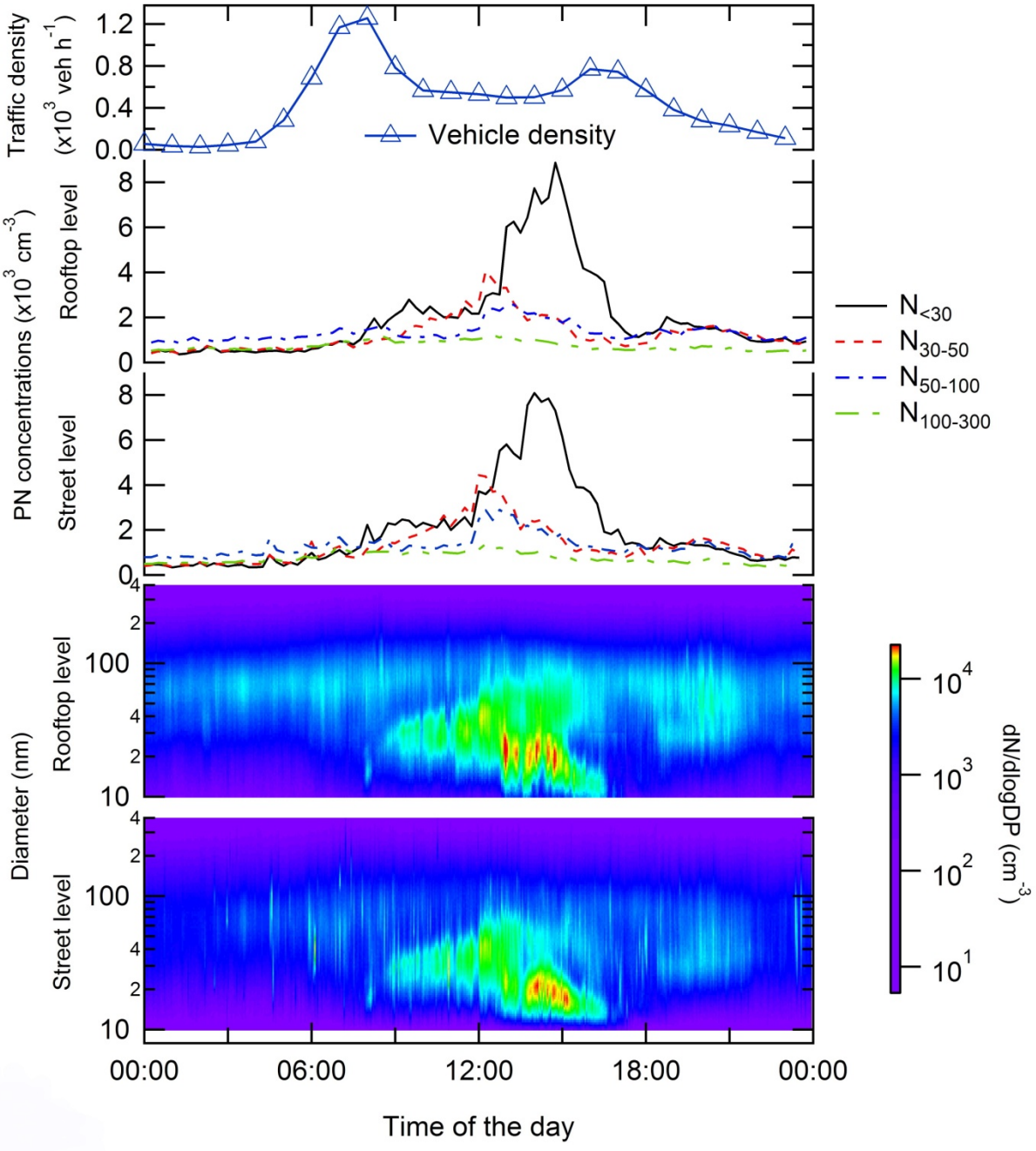


Fig. S1. Daily variation of PNSD and PN size fraction concentrations at Building B.

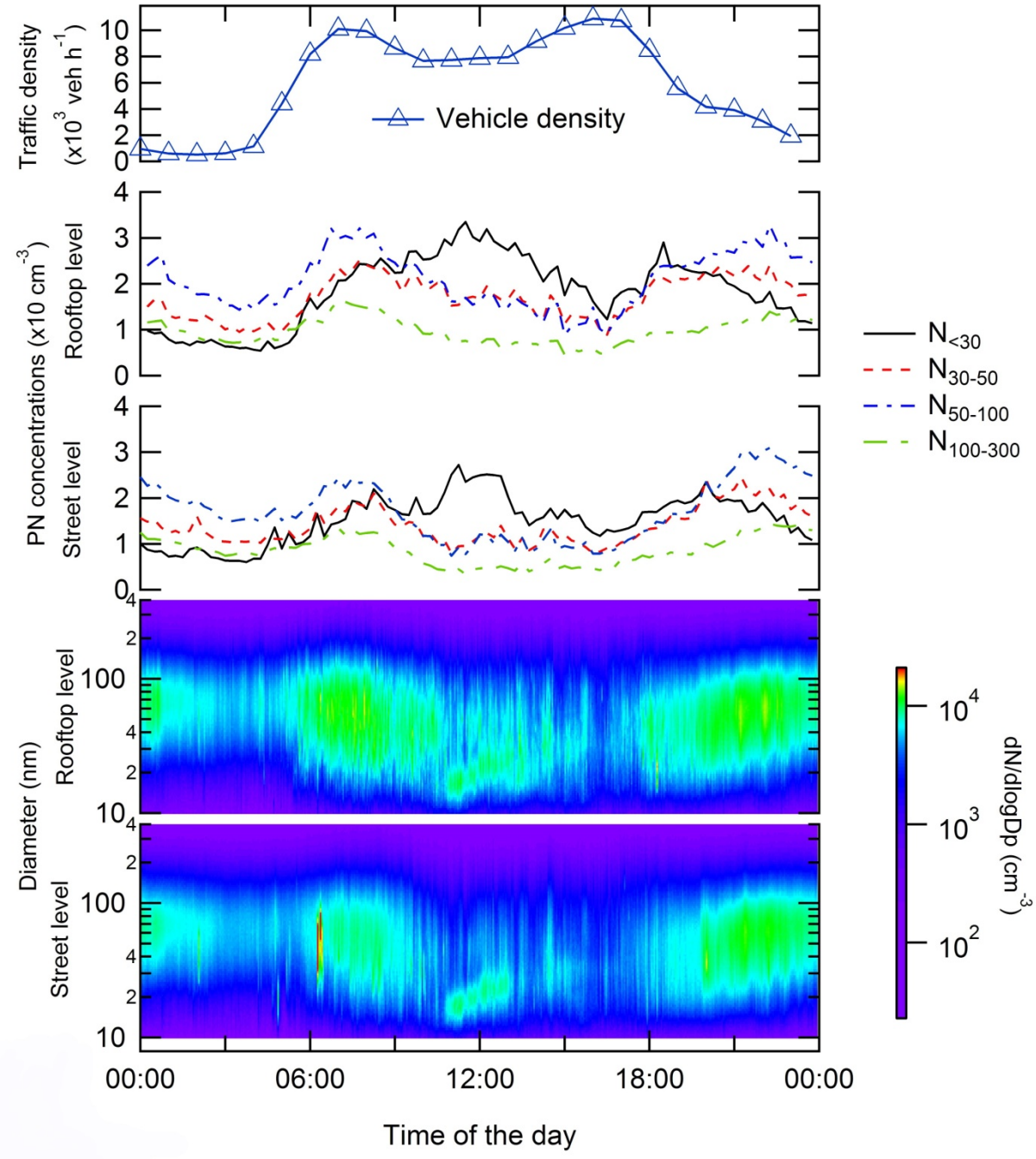
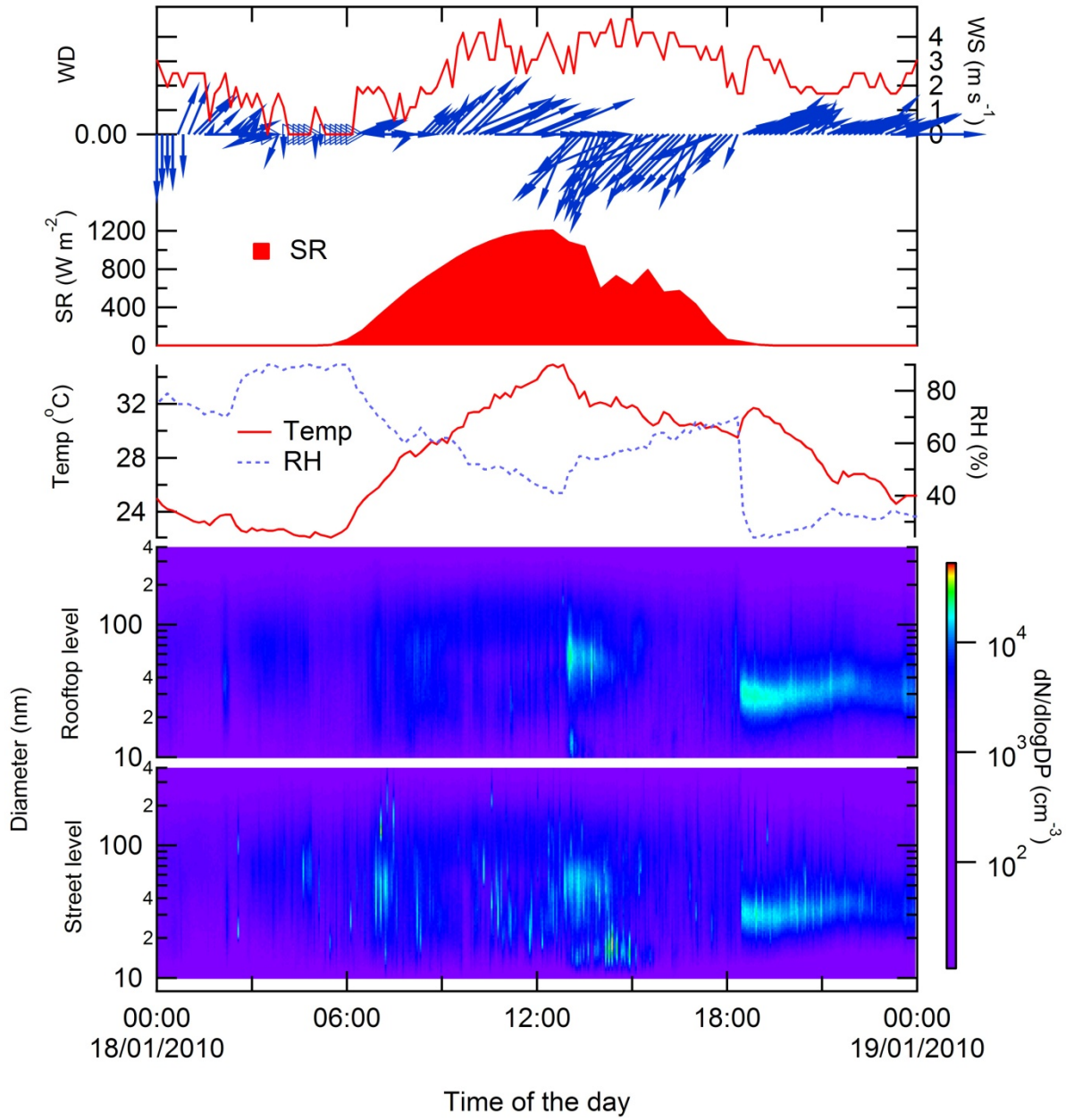


Fig. S2. Daily variation of PNSD and PN size fraction concentrations at Building C.

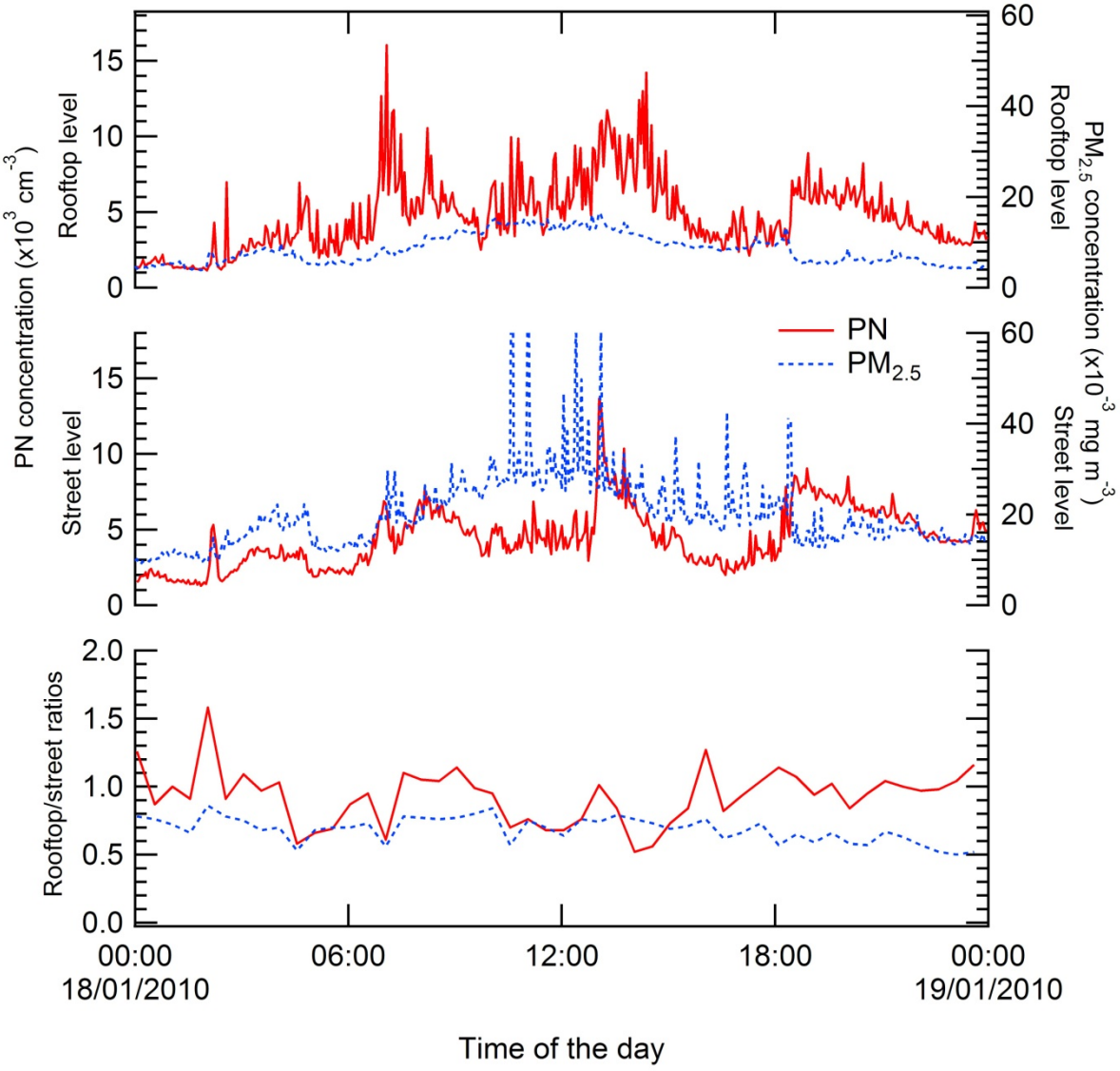


1

2 **Fig. S3.** PNSD spectra at Building B on a week day characterised by the non- or unclear

3 nucleation events.

4



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2 **Fig. S4.** Average particle concentrations and their rooftop to street level ratios at Building B

3 on a weekday characterised by the non- or unclear nucleation events.

4

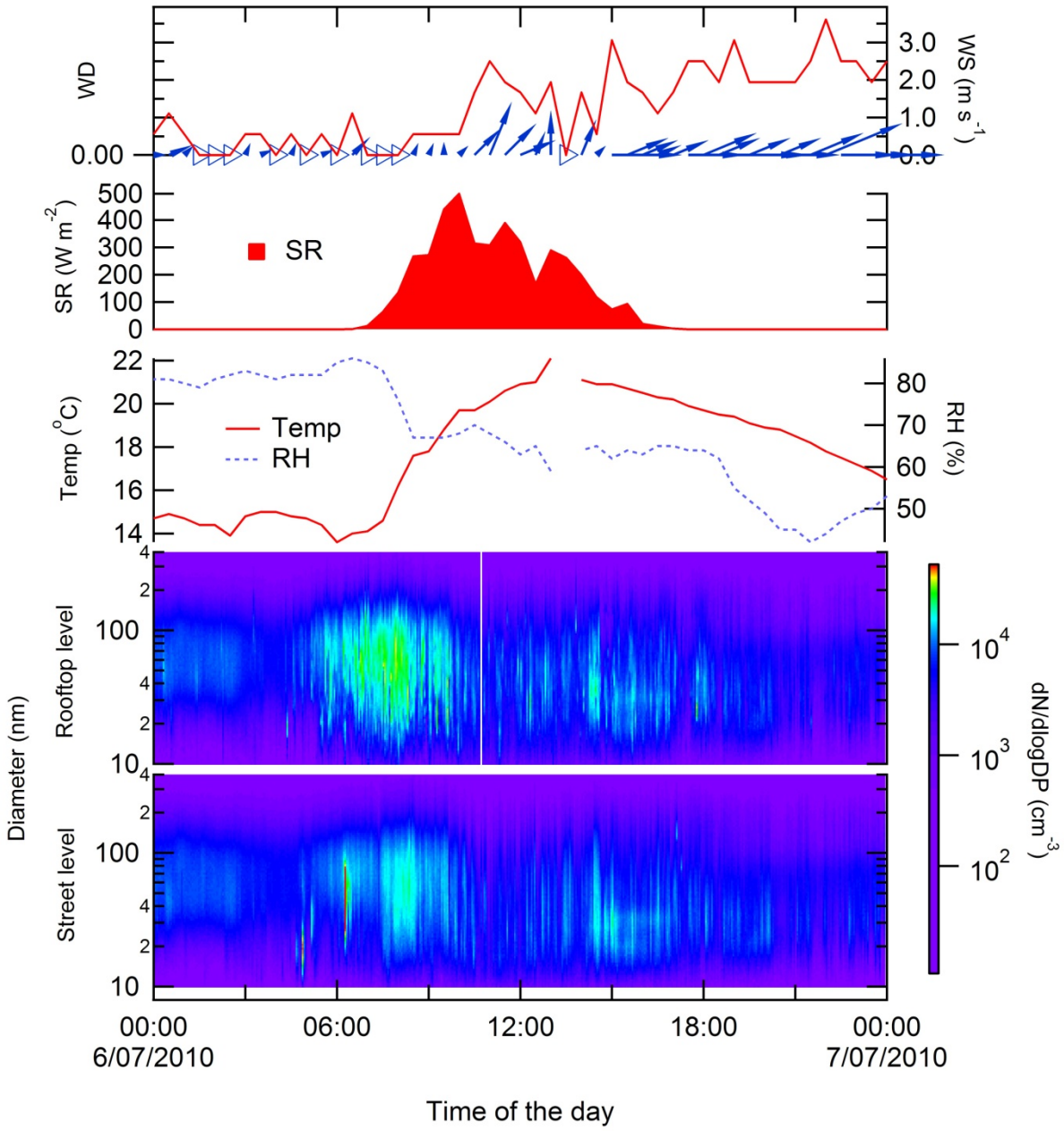
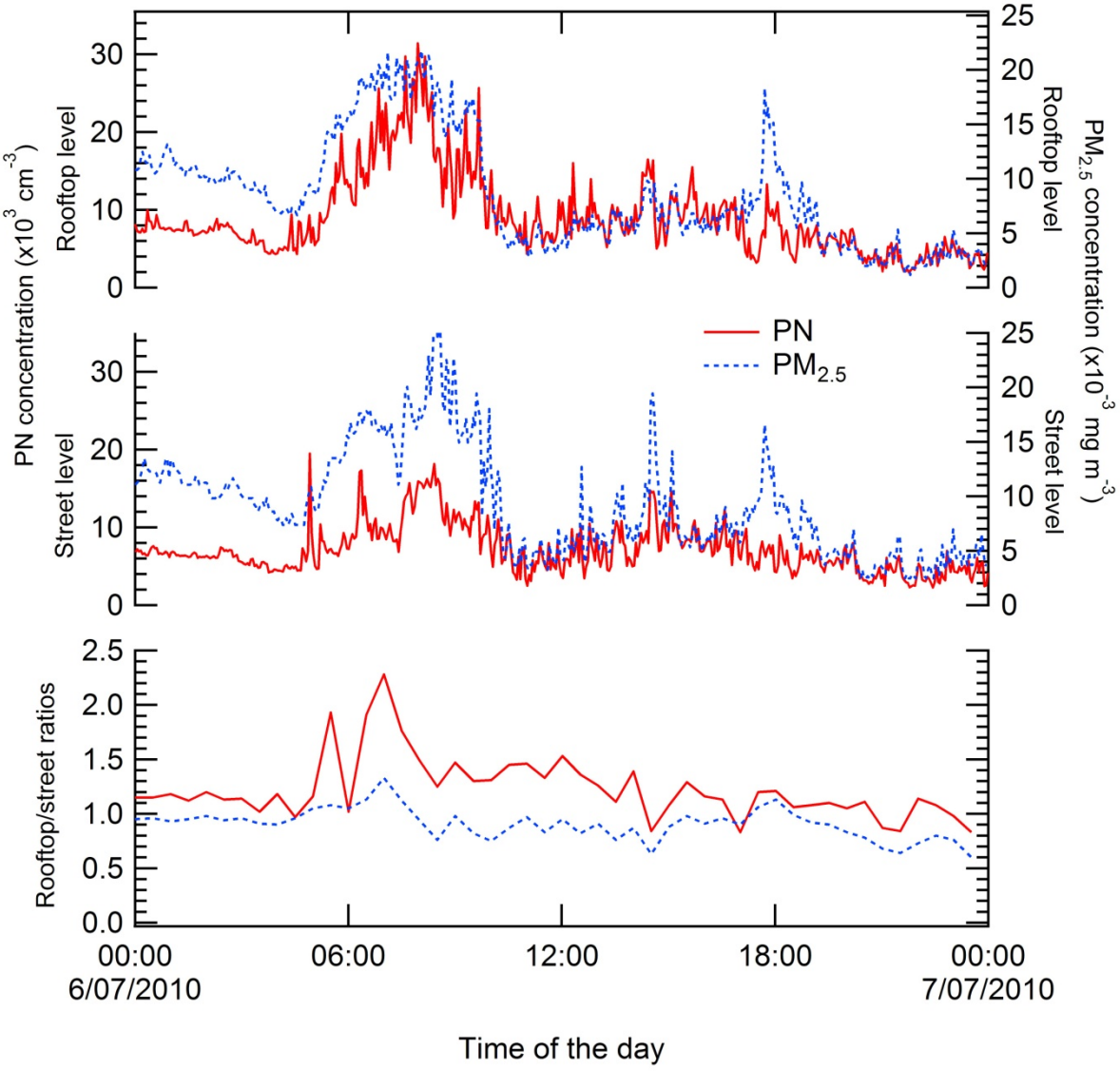


Fig. S5. PNSD spectra at Building C on a weekday characterised by the non- or unclear nucleation events.



1

2 **Fig. S6.** Average particle concentrations and their rooftop to street level ratios at Building C

3 on a weekday characterised by the non- or unclear nucleation events.

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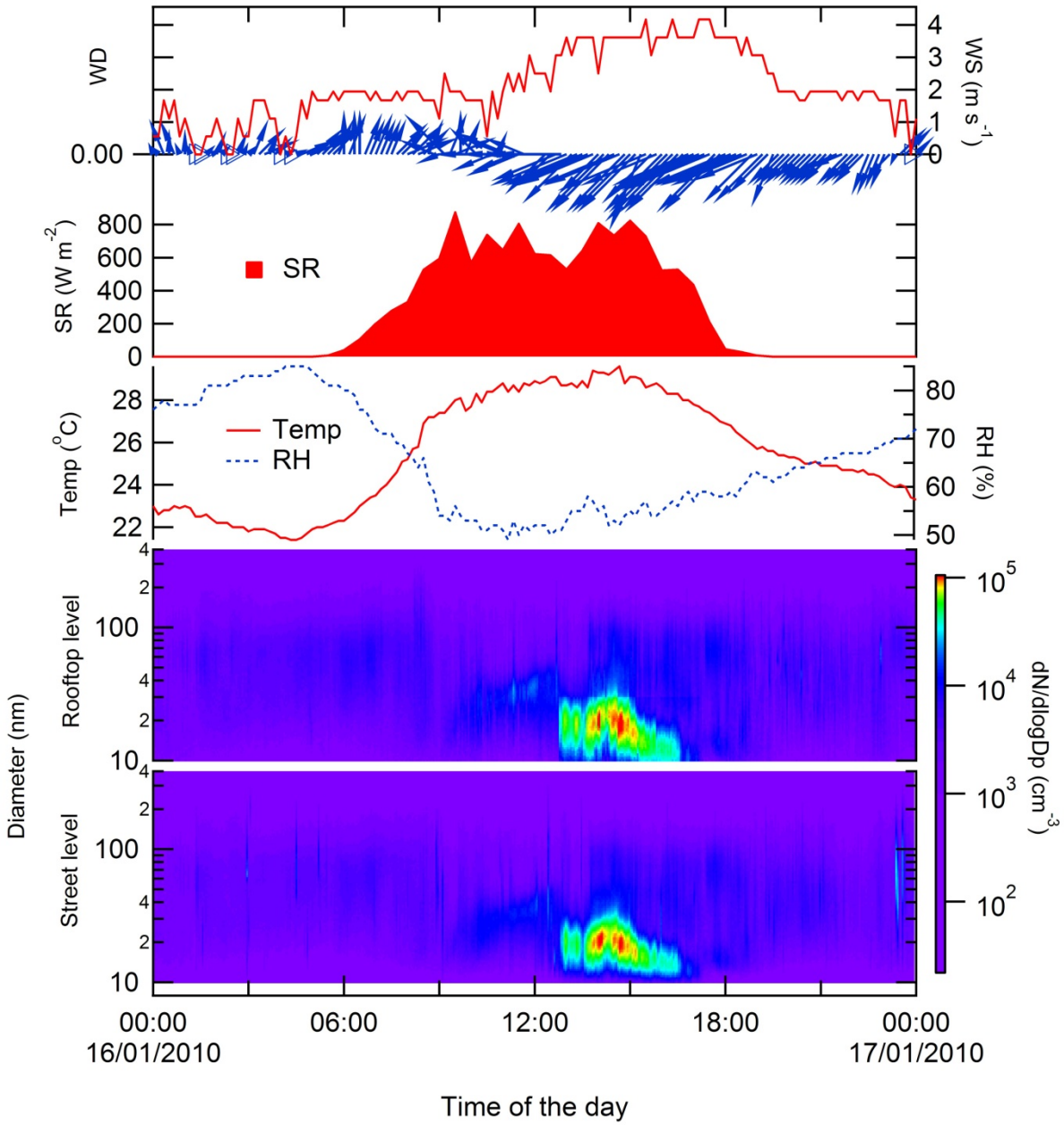

Fig. S7. PNSD spectra at Building B on a nucleation event day.

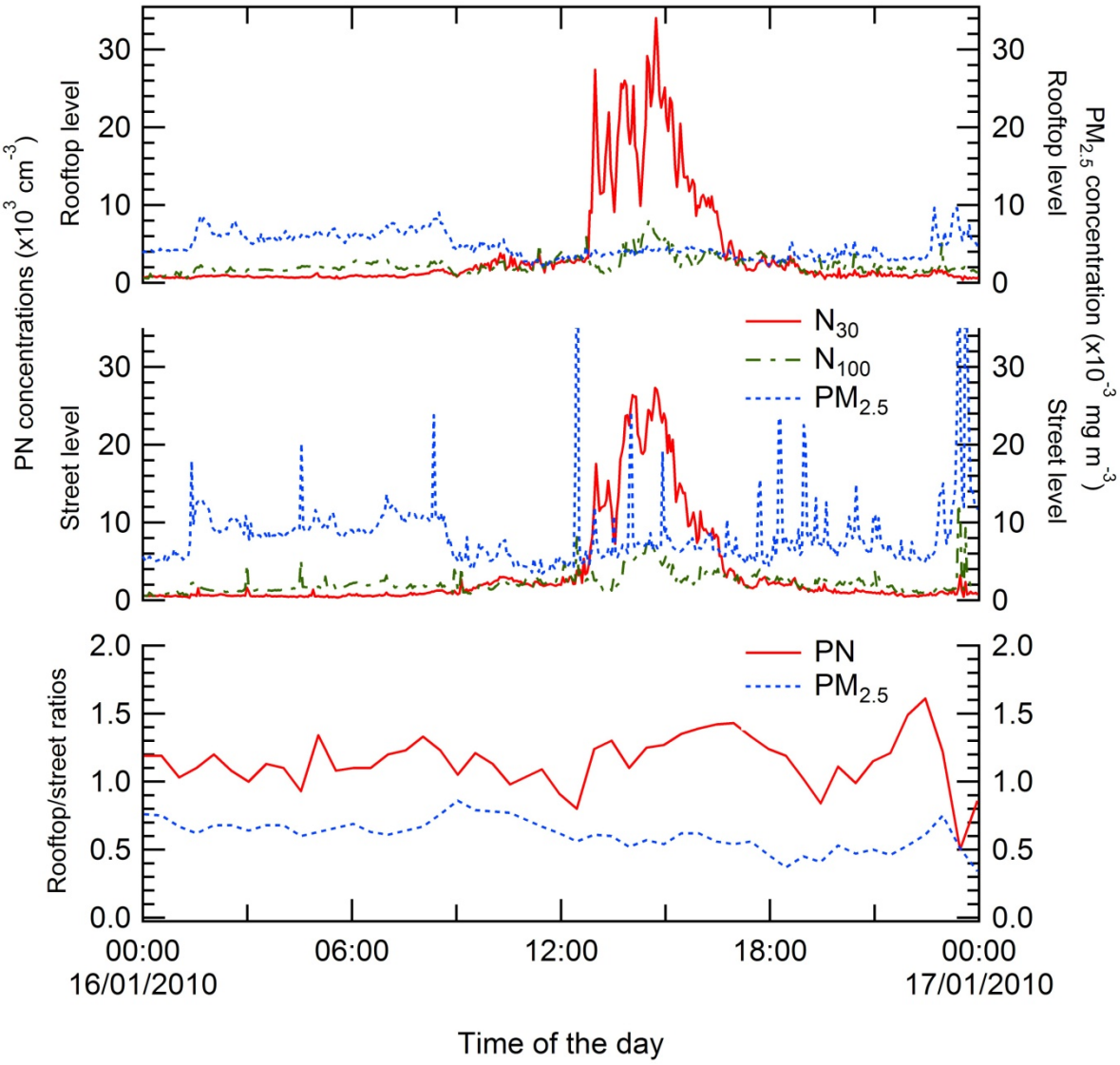

Fig. S8. Particle concentrations and their rooftop to street level ratios at Building B on a nucleation event day.

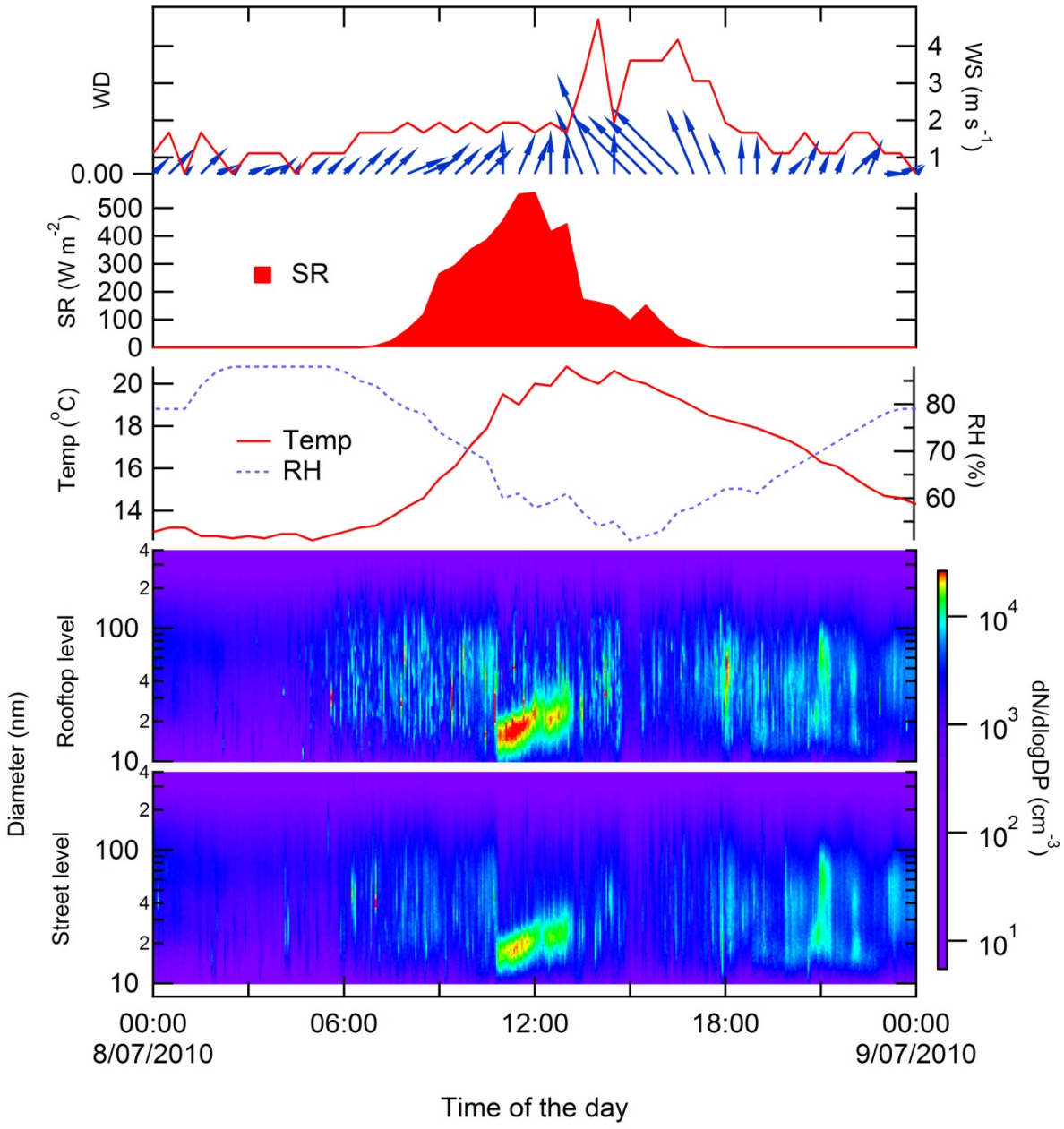

Fig. S9. PNSD spectra at Building C on a nucleation event day.

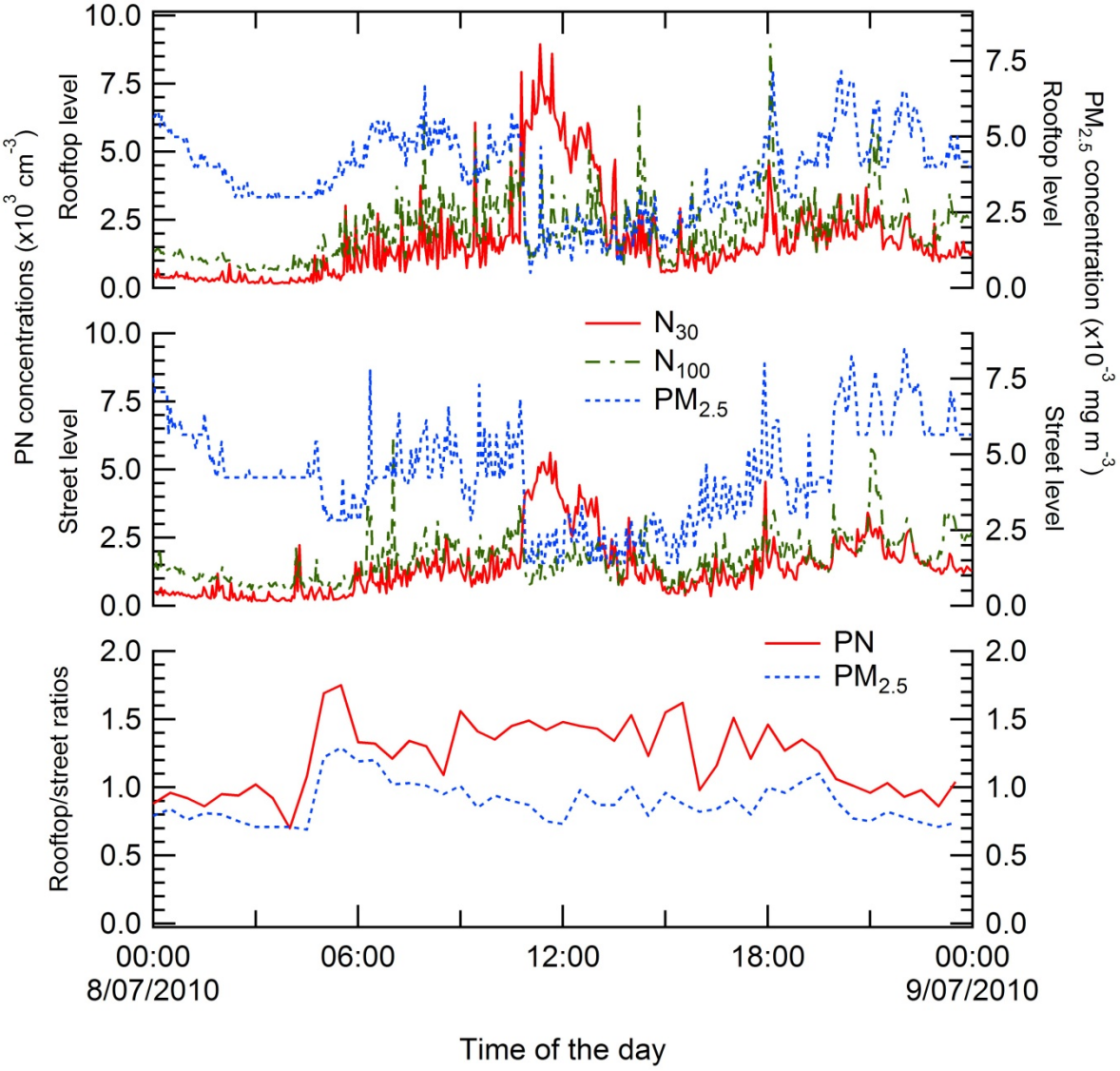

Fig. S10. Particle concentrations and their rooftop to street level ratios at Building C on a nucleation event day.

Table 1. Average meteorological conditions (± standard deviation)

Meteorological parameters	Building A 22 July – 16 August 2009	Building B 14 – 30 January 2010	Building C 24 June – 16 July 2010
Wind speed (m s^{-1})	1.7 ± 1.2	2.4 ± 1.3	1.3 ± 1.1
Solar radiation intensity (W m^{-2})	204 ± 209	343 ± 429	123 ± 203
Temperature (^{o}C)	15.7 ± 4.4	26.6 ± 3.2	15.2 ± 3.4
Relative humidity (%)	68.9 ± 18.8	63.7 ± 13.8	69.6 ± 13.1

Table 2. Average particle concentrations at the rooftop and the street levels of Buildings A, B and C during the rush-hours.

Site	Level	PN (Mean ± 95% CI) × 10^3 (cm^{-3})			$PM_{2.5}$ (Mean ± 95% CI) (μg m^{-3})		
		Morning	Afternoon	*p*	Morning	Afternoon	*p*
Building A	Rooftop	18.73 ± 1.21	9.99 ± 0.73	< 0.01	42.90 ± 1.74	10.10 ± 0.62	< 0.01
	Street	14.51 ± 0.85	7.56 ± 0.43	< 0.01	78.50 ± 3.69	11.80 ± 0.86	< 0.01
	p	< 0.01	< 0.01		< 0.01	< 0.01	
Building B	Rooftop	5.01 ± 0.37	5.82 ± 0.64	< 0.05	8.51 ± 0.48	9.59 ± 0.27	< 0.01
	Street	6.04 ± 0.65	7.21 ± 0.69	< 0.05	19.64 ± 1.14	22.02 ± 1.22	< 0.01
	p	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	
Building C	Rooftop	18.64 ± 1.21	8.56 ± 0.65	< 0.01	19.00 ± 0.51	8.00 ± 0.67	< 0.01
	Street	12.48 ± 1.70	8.12 ± 0.52	< 0.01	17.70 ± 0.79	8.20 ± 0.56	< 0.01
	p	< 0.01	0.06		< 0.05	0.45	

Table 3. Average particle concentrations during the nucleation event days.

Site	Level	$N_{<30}$ (cm^{-3}) (Mean ± 95% CI) × 10^3	$N_{<30}/N_{30-300}$ (Mean ± 95% CI)	$PM_{2.5}$ (μg m^{-3}) (Mean ± 95% CI)
Building A	Rooftop	8.16 ± 1.02	1.76 ± 0.33	11.34 ± 1.11
	Street	4.57 ± 0.28	1.01 ± 0.08	19.74 ± 3.50
	p	< 0.01	< 0.01	< 0.01
Building B	Rooftop	16.90 ± 1.49	4.54 ± 0.52	4.0 ± 0.08
	Street	15.65 ± 1.47	3.92 ± 0.34	7.5 ± 0.65
	p	< 0.05	< 0.01	< 0.01
Building C	Rooftop	5.34 ± 0.45	2.23 ± 0.32	1.67 ± 0.18
	Street	3.31 ± 0.27	1.91 ± 0.24	2.01 ± 0.14
	p	< 0.01	<0.01	< 0.01

Table S1. Summary of conditions during the nucleation events*

Site	Date	Local time	SR Wm^{-2}	WD	WS ms^{-1}	Temp ^oC	RH %
Building A	25 July 2009	10:45	722	SE	4.17	19.3	46
	31 July 2009	13:45	735	W	3.61	21	32
	1 August 2009	10:45	749	SW	2.5	18.2	47
	2 August 2009	11:30	781	WSW	2.5	19.7	55
	3 August 2009	13:30	738	WNW	4.17	23.2	32
	8 August 2009	13:30	752	E	2.5	22.1	43
	9 August 2009	9:45	656	SW	2.5	15.5	53
Building B	16 January 2010	12:45	614	NE	3.61	28.7	51
	17 January 2010	8:30	709	NNE	2.5	27.4	58
	20 January 2010	12:00	1227	NE	3.06	30.6	34
	21 January 2010	11:15	1193	NE	1.94	31.4	47
	22 January 2010	10:30	410	ENE	1.94	28.2	55

	23 January 2010	14:00	1094	ENE	4.17	31	45
	24 January 2010	14:00	1100	ENE	3.61	31.4	47
	26 January 2010	10:00	1007	N	1.94	30.2	50
	27 January 2010	11:00	1116	NE	3.61	32.7	48
Building C	4 July 2010	9:30	471	SW	1.94	13.4	53
	5 July 2010	12:30	368	SSE	1.67	21.9	49
	8 July 2010	10:45	452	SSW	1.94	19.5	60

* The data in the table present the events observed on the reference sites of Buildings A, B and C.

Table S2. Spearman's correlation coefficients (rho) for PNSD and $PM_{2.5}$ concentration around the building envelopes

Site	Measured height	Time period	Spearman's correlation coefficient (rho)			
			$N_{<30}$	N_{30-100}	$N_{>100}$	TNC
Building A	1.5 m	Daily	0.05	0.68**	0.80**	0.63**
		Rush-hours	0.21	0.22	0.78**	0.24
		Nucleation	0.49	0.63*	0.66*	0.48
	6.5 m	Daily	0.04	0.85**	0.94**	0.67**
		Rush-hours	0.46*	0.66**	0.52**	0.56**
		Nucleation	0.26	0.69**	0.71**	0.69**
	10.5 m	Daily	-0.20*	0.72**	0.88**	0.29**
		Rush-hours	0.12	0.77**	0.80**	0.49**
		Nucleation	0.17	0.72**	0.36	0.18
	14.5 m	Daily	-0.11	0.84**	0.96**	0.43**
		Rush-hours	0.27	0.60**	0.67**	0.51**
		Nucleation	-0.03	0.73**	0.90**	0.39*
Building B	1.5 m	Daily	0.53**	0.69**	0.82**	0.72**
		Rush-hours	0.13	0.20	0.64**	0.38
		Nucleation	0.66**	0.65**	0.57**	0.65**
	78.5 m	Daily	0.69**	0.82**	0.89**	0.84**
		Rush-hours	0.22	0.35	0.76**	0.43*
		Nucleation	0.78**	0.85**	0.87**	0.87**
Building C	1.5 m	Daily	0.50**	0.40**	0.44**	0.45**
		Rush-hours	0.46*	0.33	0.5*	0.41*
	5.5 m	Daily	0.37*	0.74**	0.75**	0.68**
		Rush-hours	0.55**	0.57**	0.82**	0.61**
	9.5 m	Daily	0.40*	0.85**	0.9**	0.79**
		Rush-hours	0.62**	0.68**	0.68**	0.69**
	21.5 m	Daily	0.56**	0.79**	0.60**	0.74**
		Rush-hours	0.31	0.44*	0.38*	0.46*

*. Correlation is significant at the 0.05 level (2-tailed)

**. Correlation is significant at the 0.01 level (2-tailed)